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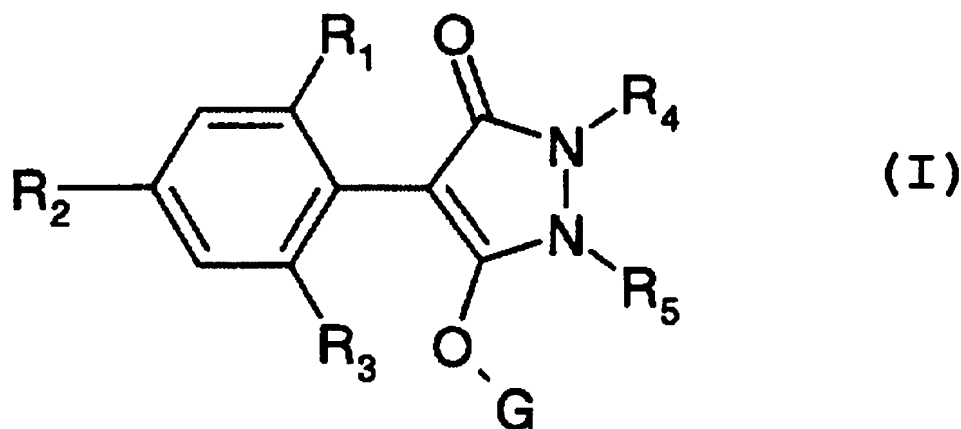
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(54) Title: NOVEL HERBICIDES



(57) Abrégé/Abstract:

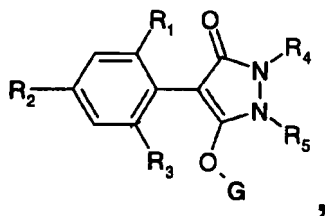
The invention relates to compounds of formula (I), wherein the substituents have the meaning cited in Claim (1). Said compounds are suitable for utilization as herbicides.



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Abstract:

Compounds of formula



wherein the substituents are as defined in claim 1,
are suitable for use as herbicides.

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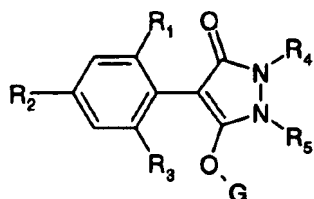
Novel herbicides

The present invention relates to novel, herbicidally active pyrazolinone derivatives, to a process for their preparation, to compositions comprising such compounds, and to the use thereof in controlling weeds, especially in crops of useful plants, or in inhibiting plant growth.

Pyrazolinone derivatives having herbicidal action are described, for example, in WO92/16510 and WO96/21652.

Novel 4-arylpyrazolinones having herbicidal and growth-inhibiting properties have now been found.

The present invention accordingly relates to compounds of formula I



I,

wherein

R₁ and R₃ are each independently of the other hydrogen, halogen, nitro, cyano, C₁-C₄alkyl, C₂-C₄alkenyl, C₂-C₄alkynyl, tri(C₁-C₄alkylsilyl)-C₂-C₄alkynyl, C₁-C₄haloalkyl, C₂-C₆haloalkenyl, C₃-C₆cycloalkyl, halo-substituted C₃-C₆cycloalkyl, benzyl, C₂-C₆alkoxyalkyl, C₂-C₆alkylthioalkyl, hydroxy, mercapto, C₁-C₆alkoxy, C₃-C₆alkenyloxy, C₃-C₆alkynyloxy, C₁-C₄alkylcarbonyl, C₁-C₄alkoxycarbonyl, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, amino, C₁-C₄alkylamino, di(C₁-C₄alkyl)amino, C₁-C₄hydroxyalkyl, formyl, C₁-C₄alkylcarbonylamino or C₁-C₄alkylsulfonylamino,

R₂ is phenyl, naphthyl or a 5- or 6-membered aromatic ring that may contain 1 or 2 hetero atoms selected from the group nitrogen, oxygen and sulfur, it being possible for the phenyl ring, the naphthyl ring and the 5- or 6-membered aromatic ring to be substituted by halogen,

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C₃-C₈cycloalkyl, hydroxy, mercapto, amino, amino-C₁-C₆alkyl, carboxyl-C₁-C₆alkyl, cyano, nitro or by formyl; and/or

for the phenyl ring, the naphthyl ring and the 5- or 6-membered aromatic ring to be substituted by C₁-C₆alkyl, C₁-C₆alkoxy, hydroxy-C₁-C₆alkyl, C₁-C₆alkoxy-C₁-C₆alkyl, C₁-C₆alkoxy-C₁-C₆alkoxy, C₁-C₆alkylcarbonyl, C₁-C₆alkylthio, C₁-C₆alkylsulfinyl, C₁-C₆alkylsulfonyl, mono-C₁-C₆alkylamino, di-C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, C₁-C₆alkylcarbonyl-(C₁-C₆alkyl)amino, C₂-C₆alkenyl, C₃-C₆alkenyloxy, hydroxy-C₃-C₆alkenyl, C₁-C₆alkoxy-C₃-C₆alkenyl, C₁-C₆alkoxy-C₃-C₆alkenyloxy, C₂-C₆alkenylcarbonyl, C₂-C₆alkenylthio, C₂-C₆alkenylsulfinyl, C₂-C₆alkenylsulfonyl, mono- or di-C₂-C₆alkenylamino, C₁-C₆alkyl-(C₃-C₆alkenyl)amino, C₂-C₆alkenylcarbonylamino, C₂-C₆alkenylcarbonyl-(C₁-C₆alkyl)amino, C₂-C₆alkynyl, C₃-C₆alkynyloxy, hydroxy-C₃-C₆alkynyl, C₁-C₆alkoxy-C₃-C₆alkynyl, C₁-C₆alkoxy-C₄-C₆alkynyloxy, C₂-C₆alkynylcarbonyl, C₂-C₆alkynylthio, C₂-C₆alkynylsulfinyl, C₂-C₆alkynylsulfonyl, mono- or di-C₃-C₆alkynylamino, C₁-C₆alkyl-(C₃-C₆alkynyl)amino, C₂-C₆alkynylcarbonylamino or by C₂-C₆alkynylcarbonyl-(C₁-C₆alkyl)amino; and/or

for the phenyl ring, the naphthyl ring and the 5- or 6-membered aromatic ring to be substituted by halo-substituted C₁-C₆alkyl, halo-substituted C₁-C₆alkoxy, halo-substituted hydroxy-C₁-C₆alkyl, halo-substituted C₁-C₆alkoxy-C₁-C₆alkyl, halo-substituted C₁-C₆alkoxy-C₁-C₆alkoxy, halo-substituted C₁-C₆alkylcarbonyl, halo-substituted C₁-C₆alkylthio, halo-substituted C₁-C₆alkylsulfinyl, halo-substituted C₁-C₆alkylsulfonyl, halo-substituted mono-C₁-C₆alkylamino, halo-substituted di-C₁-C₆alkylamino, halo-substituted C₁-C₆alkylcarbonylamino, halo-substituted C₁-C₆alkylcarbonyl-(C₁-C₆alkyl)amino, halo-substituted C₂-C₆alkenyl, halo-substituted C₃-C₆alkenyloxy, halo-substituted hydroxy-C₃-C₆alkenyl, halo-substituted C₁-C₆alkoxy-C₂-C₆alkenyl, halo-substituted C₁-C₆alkoxy-C₃-C₆alkenyloxy, halo-substituted C₂-C₆alkenylcarbonyl, halo-substituted C₂-C₆alkenylthio, halo-substituted C₂-C₆alkenylsulfinyl, halo-substituted C₂-C₆alkenylsulfonyl, halo-substituted mono- or di-C₃-C₆alkenylamino, halo-substituted C₁-C₆alkyl-(C₃-C₆alkenyl)amino, halo-substituted C₂-C₆alkenylcarbonylamino, halo-substituted C₂-C₆alkenylcarbonyl-(C₁-C₆alkyl)amino, halo-substituted C₂-C₆alkynyl, halo-substituted C₃-C₆alkynyloxy, halo-substituted hydroxy-C₃-C₆alkynyl, halo-substituted C₁-C₆alkoxy-C₃-C₆alkynyl, halo-substituted C₁-C₆alkoxy-C₄-C₆alkynyloxy, halo-substituted C₂-C₆alkynylcarbonyl, halo-substituted C₂-C₆alkynylthio, halo-substituted C₂-C₆alkynylsulfinyl, halo-substituted C₂-C₆alkynylsulfonyl, halo-substituted mono- or di-C₃-C₆alkynylamino, halo-substituted C₁-C₆alkyl-(C₃-C₆alkynyl)amino, halo-substituted C₂-C₆alkynylcarbonylamino or by halo-substituted C₂-C₆alkynylcarbonyl-(C₁-C₆alkyl)amino; and/or

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for the phenyl ring, the naphthyl ring and the 5- or 6-membered aromatic ring to be substituted by a radical of formula COOR_{50} , CONR_{51} , $\text{SO}_2\text{NR}_{53}\text{R}_{54}$ or $\text{SO}_2\text{OR}_{55}$, wherein R_{50} , R_{51} , R_{52} , R_{53} , R_{54} and R_{55} are each independently of the others hydrogen, $\text{C}_1\text{-C}_6$ alkyl, $\text{C}_2\text{-C}_6$ alkenyl or $\text{C}_3\text{-C}_6$ alkynyl, or $\text{C}_1\text{-C}_6$ alkyl, $\text{C}_2\text{-C}_6$ alkenyl or $\text{C}_3\text{-C}_6$ alkynyl each substituted by halogen, hydroxy, alkoxy, mercapto, amino, cyano, nitro, alkylthio, alkylsulfinyl or by alkylsulfonyl,

R_4 and R_5 are each independently of the other hydrogen, $\text{C}_1\text{-C}_{12}$ alkyl, $\text{C}_1\text{-C}_{12}$ haloalkyl, $\text{C}_1\text{-C}_{12}$ hydroxyalkyl, $\text{C}_3\text{-C}_8$ alkenyl, $\text{C}_3\text{-C}_8$ alkynyl, $\text{C}_1\text{-C}_{10}$ alkoxy- $\text{C}_1\text{-C}_8$ alkyl, or $\text{C}_3\text{-C}_8$ alkyl that may contain one or two oxygen atoms, $\text{C}_1\text{-C}_{10}$ alkylthio- $\text{C}_1\text{-C}_8$ alkyl, $\text{C}_3\text{-C}_8$ cycloalkyl, $\text{C}_3\text{-C}_8$ cycloalkyl that contains 1 or 2 hetero atoms selected from the group oxygen and sulfur, $\text{C}_3\text{-C}_8$ halocycloalkyl, $\text{C}_3\text{-C}_8$ halocycloalkyl that contains 1 or 2 hetero atoms selected from the group oxygen and sulfur, phenyl, or phenyl substituted by halogen, $\text{C}_1\text{-C}_6$ alkyl, $\text{C}_1\text{-C}_6$ haloalkyl, $\text{C}_1\text{-C}_6$ alkoxy, $\text{C}_1\text{-C}_6$ haloalkoxy, nitro or by cyano, or R_4 and R_5 are each independently of the other a 5- or 6-membered ring that may contain hetero atoms selected from the group oxygen, sulfur and nitrogen, or

R_4 and R_5 , together with the atoms to which they are bonded, form a 5- to 8-membered ring, which may contain 1 or 2 oxygen atoms, sulfur atoms or NR_6 groups, wherein

R_6 is hydrogen, $\text{C}_1\text{-C}_4$ alkyl, $\text{C}_1\text{-C}_6$ alkylcarbonyl, $\text{C}_1\text{-C}_6$ alkylsulfonyl, $\text{C}_3\text{-C}_6$ alkenyl or $\text{C}_3\text{-C}_6$ alkynyl, and which may be substituted by halogen, hydroxy, $\text{C}_1\text{-C}_{10}$ alkyl, $\text{C}_1\text{-C}_{10}$ alkoxy, $\text{C}_1\text{-C}_{10}$ haloalkyl, $\text{C}_3\text{-C}_8$ cycloalkyl, phenyl or by benzyl; or

which may be substituted by phenyl substituted by halogen, $\text{C}_1\text{-C}_6$ alkyl, $\text{C}_1\text{-C}_6$ haloalkyl, $\text{C}_3\text{-C}_6$ cycloalkyl, hydroxy, $\text{C}_1\text{-C}_6$ alkoxy, $\text{C}_1\text{-C}_6$ alkoxy- $\text{C}_1\text{-C}_6$ alkoxy, $\text{C}_1\text{-C}_6$ haloalkoxy or by nitro, or by benzyl substituted by halogen, $\text{C}_1\text{-C}_6$ alkyl, $\text{C}_1\text{-C}_6$ haloalkyl, $\text{C}_3\text{-C}_6$ cycloalkyl, hydroxy, $\text{C}_1\text{-C}_6$ alkoxy, $\text{C}_1\text{-C}_6$ haloalkoxy or by nitro; or

which may be substituted by CH_2 -heteroaryl, wherein the aryl moiety has 5 or 6 members, or by halo-, $\text{C}_1\text{-C}_6$ alkyl-, $\text{C}_1\text{-C}_6$ haloalkyl-, $\text{C}_1\text{-C}_6$ cycloalkyl-, hydroxy-, $\text{C}_1\text{-C}_6$ alkoxy-, $\text{C}_1\text{-C}_6$ haloalkoxy- or nitro-substituted CH_2 -heteroaryl, wherein the aryl moiety has 5 or 6 members; or which may be substituted by heteroaryl, wherein the aryl moiety has 5 or 6 members, or by halo-, $\text{C}_1\text{-C}_6$ alkyl-, $\text{C}_1\text{-C}_6$ haloalkyl-, hydroxy-, $\text{C}_1\text{-C}_6$ alkoxy-, $\text{C}_1\text{-C}_6$ haloalkoxy-, cycloalkyl- or nitro-substituted heteroaryl, wherein the aryl moiety has 5 or 6 members; and

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which may contain a fused or spiro-bound alkylene or alkenylene chain containing from 2 to 6 carbon atoms, which chain may be interrupted by oxygen or sulfur atoms,

G is hydrogen, $-C(X_1)-R_{30}$, $-C(X_2)-X_3-R_{31}$, $-C(X_4)-N(R_{32})-R_{33}$, $-SO_2-R_{34}$, an alkali metal cation, alkaline earth metal cation, sulfonium cation or ammonium cation, $-P(X_5)(R_{35})-R_{36}$ or $-CH_2X_6C(X_7)-R_{37}$, $-CH_2X_8C(X_9)-X_{10}-R_{38}$, $-CH_2X_{11}C(X_{12})-N(R_{39})-R_{40}$ or $-CH_2X_{13}SO_2-R_{41}$, wherein $X_1, X_2, X_3, X_4, X_5, X_6, X_7, X_8, X_9, X_{10}, X_{11}, X_{12}$ and X_{13} are each independently of the others oxygen or sulfur, and $R_{30}, R_{31}, R_{32}, R_{33}, R_{34}, R_{35}, R_{36}, R_{37}, R_{38}, R_{39}, R_{40}$ and R_{41} are each independently of the others hydrogen, C_1-C_{12} alkyl or C_1-C_{12} alkyl substituted by halogen, formyl, cyano, nitro, tri- C_1-C_6 alkylsilyl, hydroxy, C_1-C_6 alkoxy, C_1-C_6 alkoxycarbonyl, amino, C_1-C_6 alkylamino, di- C_1-C_6 alkylamino, mercapto, C_1-C_6 alkylthio, C_1-C_6 alkylcarbonyl, C_1-C_6 alkylcarbonylthio, C_1-C_6 alkylcarbonylamino, C_1-C_6 alkoxycarbonylamino, C_1-C_6 alkylaminocarbonylamino, C_1-C_6 alkylthiocarbonyloxy, C_1-C_6 alkylthiocarbonylamino, C_1-C_6 alkoxythiocarbonyl, aminothiocarbonyl, C_1-C_6 alkylthiocarbonyloxy, C_1-C_6 alkylthiocarbonylamino, C_1-C_6 alkoxythiocarbonylamino, C_1-C_6 alkylsulfinyl, C_1-C_6 alkylsulfonyl, C_1-C_6 alkylsulfonyloxy, C_1-C_6 alkylsulfonylamino, C_1-C_6 alkoxyimino, hydroxyimino, heteroaryl, benzyloxy, phenoxy or by halophenoxy; or

C_2-C_{12} alkenyl, C_3-C_{12} cycloalkyl, C_3-C_6 cycloalkyl substituted by halogen, C_1-C_6 haloalkyl, C_1-C_6 alkyl, C_1-C_6 alkoxy, C_1-C_6 alkylcarbonyloxy, C_1-C_6 thioalkyl, C_1-C_6 alkylcarbonylthio, C_1-C_6 alkylamino, C_1-C_6 alkylcarbonylamino, tri- C_1-C_6 alkylsilyl or by tri- C_1-C_6 alkylsilyloxy; phenyl or phenyl substituted by alkoxy, halogen, C_1-C_6 haloalkyl, nitro, cyano, C_1-C_6 alkyl, C_1-C_6 alkylcarbonyloxy, C_1-C_6 thioalkyl, C_1-C_6 alkylcarbonylthio, C_1-C_6 alkylamino, C_1-C_6 alkylcarbonylamino, tri- C_1-C_6 alkylsilyl or by tri- C_1-C_6 alkylsilyloxy; heteroaryl or heteroaryl substituted by halogen, C_1-C_6 haloalkyl, nitro, cyano, C_1-C_6 alkyl, C_1-C_6 alkoxy, C_1-C_6 alkylcarbonyloxy, C_1-C_6 thioalkyl, C_1-C_6 alkylcarbonylthio, C_1-C_6 alkylamino, C_1-C_6 alkylcarbonylamino, tri- C_1-C_6 alkylsilyl or by tri- C_1-C_6 alkylsilyloxy; and

R_{34} is additionally C_2-C_{20} alkenyl or C_2-C_{20} alkenyl substituted by halogen, C_1-C_6 alkylcarbonyl, C_1-C_6 alkoxycarbonyl, C_1-C_6 alkylcarbonyloxy, C_1-C_6 alkoxy, C_1-C_6 thioalkyl, C_1-C_6 alkylthiocarbonyl, C_1-C_6 alkylcarbonylthio, C_1-C_6 alkylsulfonyl, C_1-C_6 alkylsulfinyl, C_1-C_6 alkylaminosulfonyl, di- C_1-C_6 alkylaminosulfonyl, C_1-C_6 alkylsulfonyloxy, C_1-C_6 alkylsulfonylamino, C_1-C_6 alkylamino, di- C_1-C_6 alkylamino, C_1-C_6 alkylcarbonylamino, di- C_1-C_6 alkylcarbonylamino, cyano, C_3-C_6 cycloalkyl, C_3-C_6 heterocyclyl, tri- C_1-C_6 alkylsilyl, tri- C_1-C_6 alkylsilyloxy, phenyl, substituted phenyl, heteroaryl or by substituted heteroaryl; or

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C₂-C₂₀alkynyl or C₂-C₂₀alkynyl substituted by halogen, C₁-C₆alkylcarbonyl, C₁-C₆alkoxy-carbonyl, C₁-C₆alkylcarbonyloxy, C₁-C₆alkoxy, C₁-C₆thioalkyl, C₁-C₆alkylthiocarbonyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylsulfonyl, C₁-C₆alkylsulfinyl, C₁-C₆alkylaminosulfonyl, di-C₁-C₆alkylaminosulfonyl, C₁-C₆alkylsulfonyloxy, C₁-C₆alkylsulfonylamino, C₁-C₆alkylamino, di-C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, di-C₁-C₆alkylcarbonylamino, cyano, C₃-C₇-cycloalkyl, C₃-C₇heterocyclyl, tri-C₁-C₆alkylsilyl, tri-C₁-C₆alkylsilyloxy, phenyl, substituted phenyl, heteroaryl or by substituted heteroaryl; or

C₃-C₆cycloalkyl or C₃-C₆cycloalkyl substituted by halogen, C₁-C₆haloalkyl, C₁-C₆alkyl, C₁-C₆-alkoxy, C₁-C₆alkylcarbonyloxy, C₁-C₆thioalkyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, tri-C₁-C₆alkylsilyl or by tri-C₁-C₆alkylsilyloxy; or

heteroaryl or heteroaryl substituted by halogen, C₁-C₆haloalkyl, nitro, cyano, C₁-C₆alkyl, C₁-C₆alkoxy, C₁-C₆alkylcarbonyloxy, C₁-C₆thioalkyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, tri-C₁-C₆alkylsilyl or by tri-C₁-C₆alkylsilyloxy; or

heteroaryloxy, substituted heteroaryloxy, heteroarylthio, substituted heteroarylthio, heteroarylamino, substituted heteroarylamino, diheteroarylamino, substituted diheteroarylamino, phenylamino, substituted phenylamino, diphenylamino, substituted diphenylamino, cycloalkylamino, substituted cycloalkylamino, dicycloalkylamino, substituted dicycloalkylamino, cycloalkoxy or substituted cycloalkoxy,

and to salts and diastereoisomers of the compounds of formula I.

The alkyl groups occurring in the definitions of the substituents may be straight-chained or branched and are, for example, methyl, ethyl, n-propyl, isopropyl, n-butyl, sec-butyl, isobutyl, tert-butyl, pentyl and hexyl and branched isomers thereof. Suitable alkenyl and alkynyl groups and alkoxy and alkylthio groups and other groups containing an alkyl unit are derived from the mentioned alkyl groups. Examples of suitable cycloalkyl groups according to the invention are cyclopropyl, cyclobutyl, cyclopentyl, cyclohexyl, cycloheptyl and cyclooctyl. Halogen substituents are preferably fluorine, chlorine or bromine. Examples of 5- and 6-membered aromatic rings having hetero atoms are thienyl, furyl and pyridyl. As 5- to 8-membered heteroaryls and heterocycloalkyls there may be mentioned, for example, pyrazolidine, 1,2,3,6-tetrahydropyridazine, hexahydropyridazine, 1,4,5-oxadiazepane, 1,4,5-thiadiazepane and 1,4,5-oxadiazoxane.

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The invention relates also to the salts that the compounds of formula I can form preferably with amines, alkali metal and alkaline earth metal bases or with quaternary ammonium bases. Suitable salt formers are described, for example, in WO 98/41089.

The invention relates also to the salts that the compounds of formula I can form with amines, alkali metal and alkaline earth metal bases or with quaternary ammonium bases.

Among the alkali metal and alkaline earth metal hydroxides as salt formers, special mention may be made of the hydroxides of lithium, sodium, potassium, magnesium or calcium, but especially those of sodium or potassium.

As examples of amines suitable for ammonium salt formation there come into consideration both ammonia and primary, secondary and tertiary C₁-C₁₈alkylamines, C₁-C₄hydroxyalkylamines and C₂-C₄alkoxyalkylamines, for example methylamine, ethylamine, n-propylamine, isopropylamine, the four isomeric butylamines, n-amylamine, isoamylamine, hexylamine, heptylamine, octylamine, nonylamine, decylamine, pentadecylamine, hexadecylamine, heptadecylamine, octadecylamine, methyl-ethylamine, methyl-isopropylamine, methyl-hexylamine, methyl-nonylamine, methyl-pentadecylamine, methyl-octadecylamine, ethyl-butylamine, ethyl-heptylamine, ethyl-octylamine, hexyl-heptylamine, hexyl-octylamine, dimethylamine, diethylamine, di-n-propylamine, diisopropylamine, di-n-butylamine, di-n-amylamine, diisoamylamine, dihexylamine, diheptylamine, dioctylamine, ethanolamine, n-propanolamine, isopropanolamine, N,N-diethanolamine, N-ethylpropanolamine, N-butylethanolamine, allylamine, n-butenyl-2-amine, n-pentenyl-2-amine, 2,3-dimethylbutenyl-2-amine, di-butenyl-2-amine, n-hexenyl-2-amine, propylenediamine, trimethylamine, triethylamine, tri-n-propylamine, triisopropylamine, tri-n-butylamine, triisobutylamine, tri-sec-butylamine, tri-n-amylamine, methoxyethylamine and ethoxyethylamine; heterocyclic amines, such as pyridine, quinoline, isoquinoline, morpholine, piperidine, pyrrolidine, indoline, quinuclidine and azepine; primary arylamines, such as anilines, methoxyanilines, ethoxyanilines, o-, m- and p-toluidines, phenylenediamines, benzidines, naphthylamines and o-, m- and p-chloroanilines; but especially triethylamine, isopropylamine and diisopropylamine.

Preferred quaternary ammonium bases suitable for salt formation correspond, for example, to the formula $[N(R_a R_b R_c R_d)]OH$, wherein R_a, R_b, R_c and R_d are each independently of the

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others C₁-C₄alkyl. Other suitable tetraalkylammonium bases with other anions can be obtained, for example, by anion exchange reactions.

Preferred compounds of formula I are those wherein R₁ and R₃ are each independently of the other C₁-C₄alkyl, especially methyl or ethyl, C₂-C₄alkynyl, especially ethynyl, C₁-C₆alkoxy, especially methoxy, C₁-C₄alkylthio, especially methylthio, C₁-C₄haloalkyl, especially chloromethyl and chloroethyl, formyl, C₁-C₄alkylcarbonyl, especially acetyl, or C₁-C₄alkylamino or di(C₁-C₄)alkylamino. Special preference is given to methyl, ethyl, ethynyl and methoxy.

In a further group of preferred compounds of formula I, R₂ is phenyl, 2-thienyl, 3-thienyl, 2-furyl, 3-furyl, 2-pyridyl, 3-pyridyl or 4-pyridyl, with phenyl being especially preferred. Preference is given also to the corresponding substituted ring systems, with halogen, hydroxy, mercapto, amino, cyano, nitro, formyl, C₁-C₆alkyl, C₁-C₈haloalkyl and C₃-C₈cycloalkyl coming into consideration as substituents.

Other preferred compounds of formula I are those wherein R₄ and R₅ are each independently of the other C₁-C₁₂alkyl, C₁-C₁₀alkoxy-C₁-C₈alkyl or C₃-C₈alkyl that may contain one or two oxygen atoms, or R₄ and R₆, together with the atoms to which they are bonded, form a saturated 5- to 8-membered, especially 6- or 7-membered, ring. Especially preferably R₄ and R₅ together form a radical -CH₂CH₂-O-CH₂CH₂- or -(CH₂)₄-, it being possible for those radicals to be substituted by C₁-C₁₀alkyl, C₁-C₁₀alkoxy, C₁-C₈alkoxy-C₁-C₆alkoxy or by hydroxyl and to carry an alkylene or alkenylene chain containing from 2 to 6 carbon atoms, which chain may be interrupted by oxygen.

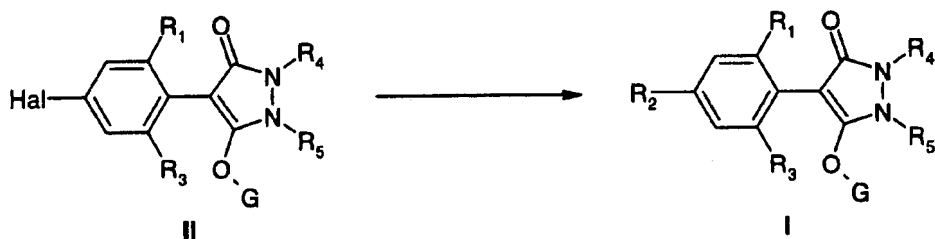
Further preferred compounds of formula I have as substituent G hydrogen or COR₃₀, especially pivaloyl, C(O)X₃R₃₁ or SO₂R₃₄, wherein X₃ is oxygen or sulfur, R₃₁ is C₁-C₁₂alkyl, C₁-C₁₂haloalkyl, C₂-C₁₂alkenyl, C₁-C₁₂alkoxyalkyl, C₃-C₁₂cycloalkyl or phenyl, and R₃₄ is C₁-C₆alkyl, C₁-C₆haloalkyl, phenyl, chloro-, cyano- or methyl-substituted phenyl; or heteroaryl or chloro-, cyano- or methyl-substituted heteroaryl.

In a further preferred group of compounds of formula I, R₁ and R₃ are each independently of the other C₁-C₄alkyl, especially methyl or ethyl, C₂-C₄alkynyl, especially ethynyl, C₁-C₆-

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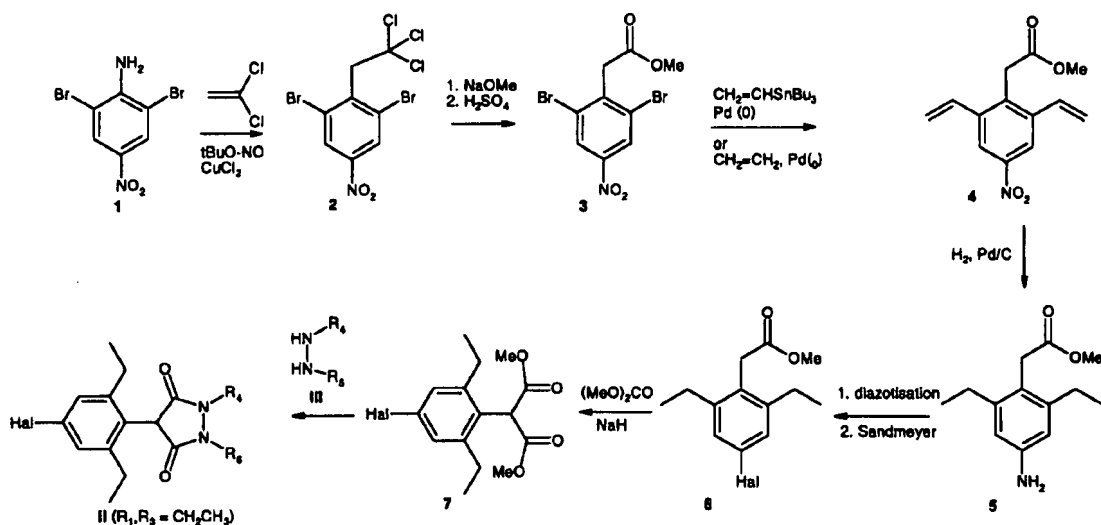
alkoxy, especially methoxy, or C₁-C₄alkylthio, especially methylthio, R₂ is phenyl, 2-thienyl, 3-thienyl, 2-furyl or 3-furyl, R₄ and R₅ are each independently of the other C₁-C₁₂alkyl, or R₄ and R₅, together with the atoms to which they are bonded, form a radical -CH₂CH₂-O-CH₂CH₂- or -(CH₂)₄-, it being possible for those radicals to be substituted by C₁-C₁₀alkyl or by C₁-C₆alkoxy-C₁-C₆alkoxy, and G is hydrogen or COR₃₀, especially pivalyl, or C(O)X₃R₃₁, wherein X₃ is oxygen and R₃₁ is C₁-C₁₂alkyl, C₃-C₁₂cycloalkyl or phenyl.

The compounds of formula I can be prepared by processes known *per se*, for example processes described in WO 97/02243 or in the literature references cited below, by, for example, introducing the aromatic ring R₂ by Pd(0)-catalysed cross-coupling of aromatic tin compounds, for example phenyl-trialkyltin compounds, (Stille, J.K. *Angew. Chem. Int.Ed. Engl.* 1986, 25, 508. Kwon, H.B.; McKee, B.H.; Stille, J.K. *J. Org. Chem.* 1990, 55, 3114), or zinc compounds, such as phenyl-zinc halides (Negishi, E.; Valente, L.F.; Kobayashi, M. *J. Am. Chem. Soc.* 1980, 102, 3298. Knochel, P.; Singer, R. *Chem. Rev.* 1993, 93, 2117), or aromatic boric acids, such as furyl-boric acids (Miyaura, N.; Yanagi, T.; Suzuki, A. *Synth. Commun.* 1981, 11, 513), or aromatic boric acid esters, such as phenyl-boric acid dialkyl esters (Sato, M.; Miyaura, N.; Suzuki, A. *Chem. Lett.* 1989, 1405. Watanabe, T.; Miyaura, N.; Suzuki, A. *Synlett* 1992, 3, 207), or aromatic Grignard compounds, such as phenyl-magnesium bromide (Jendrella, H.; Chen, I. J., *Synthesis* 1990, 827; Widdowson, D. A.; Zhang, Y. Z., *Tetrahedron* 1986, 42, 2111) with the halogen compounds II (Hal = chlorine, bromine or iodine).

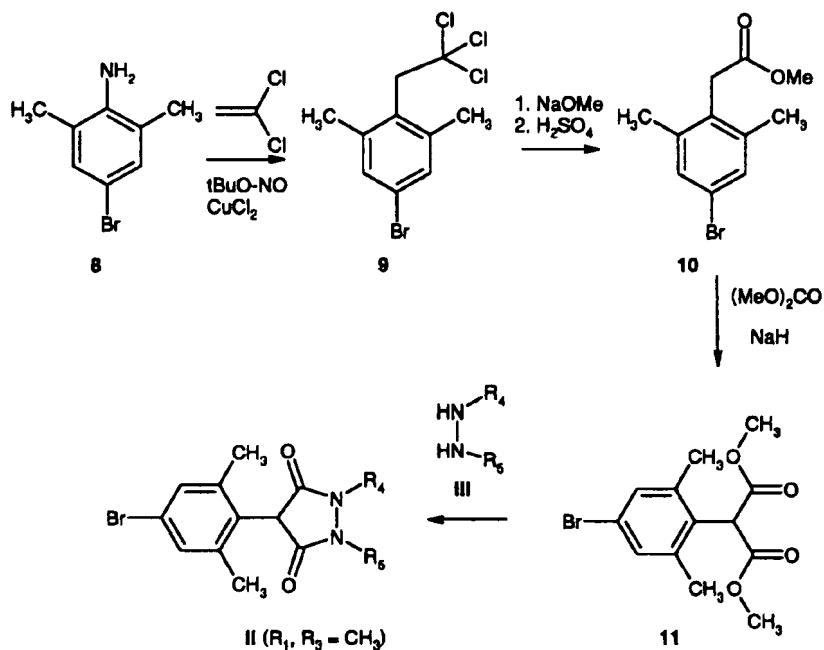


The compound of formula II can be prepared analogously as described in WO 97/02243 or, for example, according to the following Scheme for II wherein R₁, R₂ = CH₂CH₃ (Hal = Cl, Br, I)

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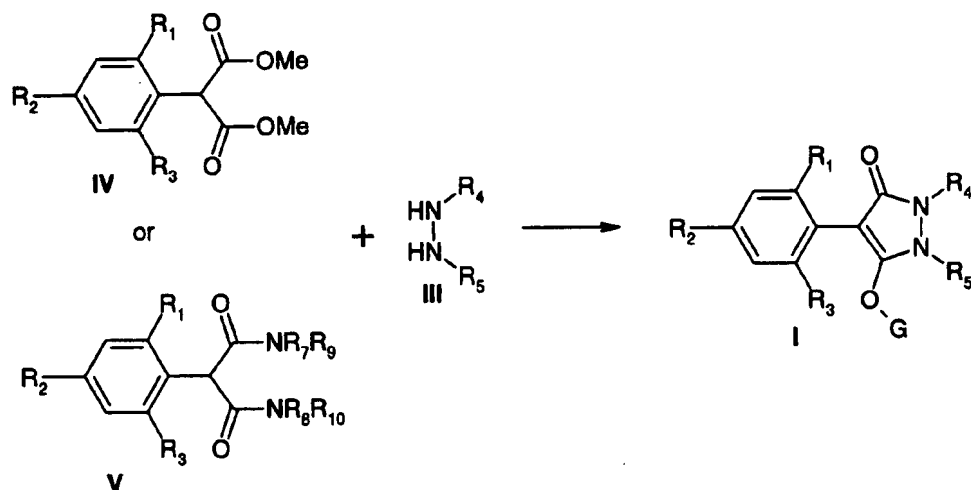


The corresponding dimethyl compound (II wherein $R_1, R_2 = \text{CH}_3$) can be prepared, for example, according to the following Scheme:

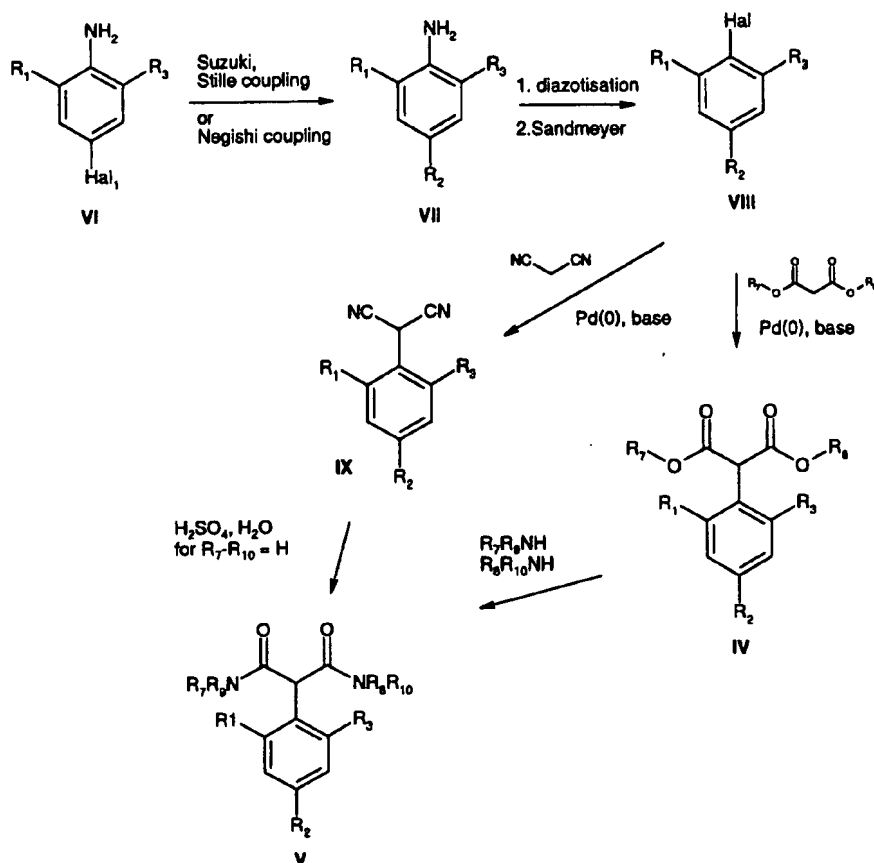


Another method of preparing the compounds according to the invention is based on coupling the malonic acid ester IV or malonodiamide V with a substituted hydrazine III. R_7 to R_{10} can then be hydrogen and/or alkyl, especially C_1 - C_6 alkyl, and/or aryl, preferably phenyl and naphthyl.

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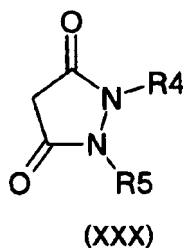
The malonic acid ester IV or the malonodiamide V can be prepared, for example, by Pd(0)-catalysed cross-coupling according to the following Scheme:



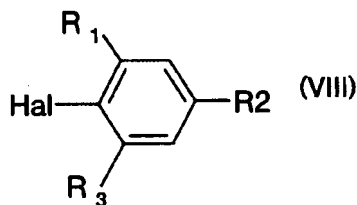
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The Suzuki, Stille or Negishi coupling of **VI** to form **VII** can be carried out according to the above-mentioned procedures ($\text{Hal}_1 = \text{Br}$ or I); diazotisation and Sandmeyer reaction (Vogel's Textbook of Practical Organic Chemistry, 5th Edition, B.S. Furniss, A.J. Hannaford, P.W.G. Smith, A.R. Tatchell; Longman Scientific & Technical 1989, page 923) result in halogen compounds **VIII** ($\text{Hal}_2 = \text{Cl}$, Br or I) which can be converted directly to the phenyl malonates **IV** by $\text{Pd}(0)$ cross-couplings (Kawatsura, M.; Hartwig, J.F. J. Am. Chem. Soc. **1999**, 121, 1473). Starting from **VIII** by means of analogous $\text{Pd}(0)$ cross-couplings, there are obtainable malonodinitriles **IX**, from which amides **V** are obtainable.

Compounds of formula I wherein G is hydrogen can also be prepared by reacting a compound of formula XXX



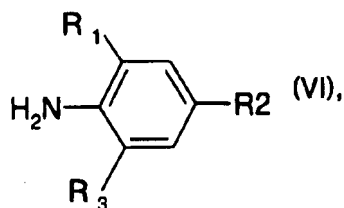
wherein R_4 and R_5 are as defined above, with a compound of formula VIII



wherein R_1 , R_2 and R_3 are as defined for formula I and Hal is chlorine, bromine or iodine, in the presence of an inert solvent, a base and a palladium catalyst at temperatures of from 30 to 250 °C. The reaction is preferably carried out under an inert gas atmosphere.

The compounds of formula XXX are known or can be prepared according to known processes, as described, for example, in J. Chem. Soc. Perkin Trans. 1 (1987), (4), 877-884. The compounds of formula VIII can be prepared, for example, according to known methods, by way of the diazonium salts e.g. Sandmeyer reaction, from the corresponding anilines of

formula VI



wherein R_1 and R_3 are as defined for formula I. Such reactions are described, for example, in Vogel's Textbook of Practical Organic Chemistry, 5th Edition, B.S. Furniss, A.J. Hannaford, P.W.G. Smith, A.R. Tatchell; Longman Scientific & Technical 1989, page 923. The compounds of formula VI are known, some of them are commercially available or they can be prepared analogously to known compounds.

Suitable for that reaction are bases, such as tri-alkali metal phosphates, alkali metal and alkaline earth metal hydrides, alkali metal and alkaline earth metal amides or alkali metal alcoholates, for example tripotassium phosphate, sodium hydride, lithium diisopropylamide (LDA), sodium tert-butanolate or potassium tert-butanolate. Special preference is given to sodium tert-butanolate, potassium tert-butanolate and tripotassium phosphate.

Suitable solvents are, for example, aromatic hydrocarbons, such as xylene or toluene, ethers, such as tetrahydrofuran, dioxane or ethylene glycol dimethyl ether, dimethyl sulfoxide or tertiary amides, such as dimethylformamide, N-methylpyrrolidinone or dimethylacetamide or acyclic ureas, such as N,N'-dimethylpropyleneurea.

The palladium catalysts suitable for the C-C linkage reaction of a compound of formula XXX with a compound of formula VIII are generally palladium(II) or palladium(0) complexes, such as palladium(II) dihalides, palladium(II) acetate, palladium(II) sulfate, bis(triphenylphosphine)palladium(II) dichloride, bis(tricyclopentylphosphine)palladium(II) dichloride, bis(tricyclohexylphosphine)palladium(II) dichloride, bis(dibenzylideneacetone)palladium(0) or tetrakis(triphenylphosphine)palladium(0). The palladium catalyst can also be prepared from palladium(II) or palladium(0) compounds by complexing with the desired ligands *in situ*, for example by placing the palladium(II) salt to be complexed, for example palladium(II) dichloride (PdCl_2) or palladium(II) acetate ($\text{Pd}(\text{OAc})_2$), together with the desired ligand, for

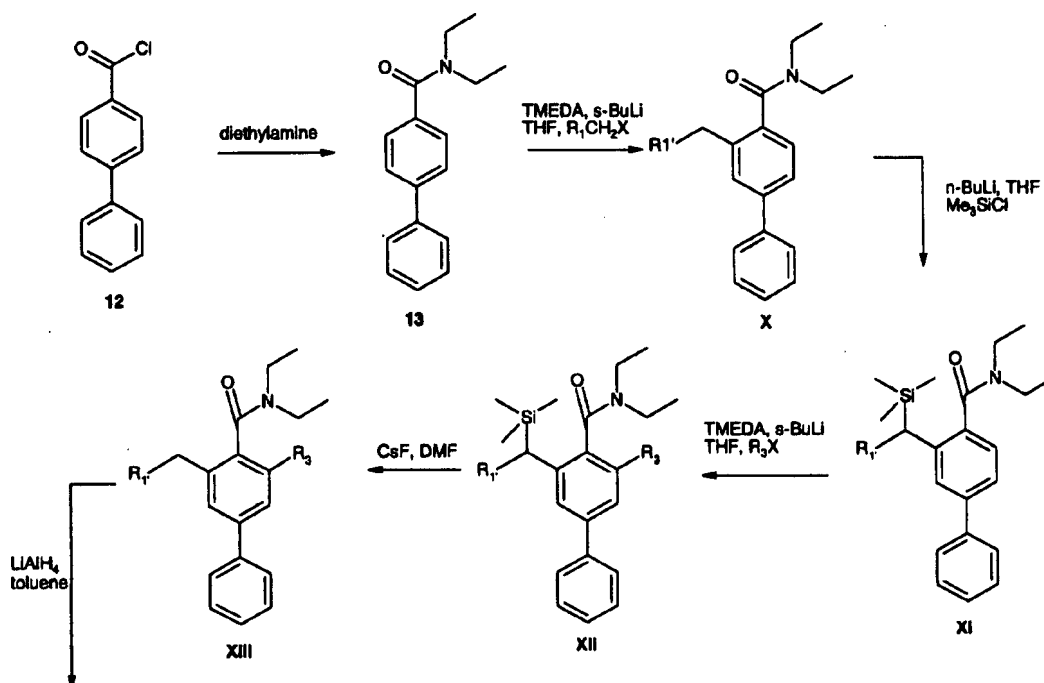
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example triphenylphosphine (PPh_3), tricyclopentylphosphine or tricyclohexylphosphine, together with the selected solvent, a compound of formula VIII, a compound of formula XXX and base. Bidendate ligands are also suitable, for example 1,1'-bis(diphenylphosphino)-ferrocene or 1,2-bis(diphenylphosphino)ethane. By heating the reaction mixture, the palladium(II) or palladium(0) complex desired for the C-C coupling reaction is formed *in situ*, and said complex then initiates the C-C coupling reaction.

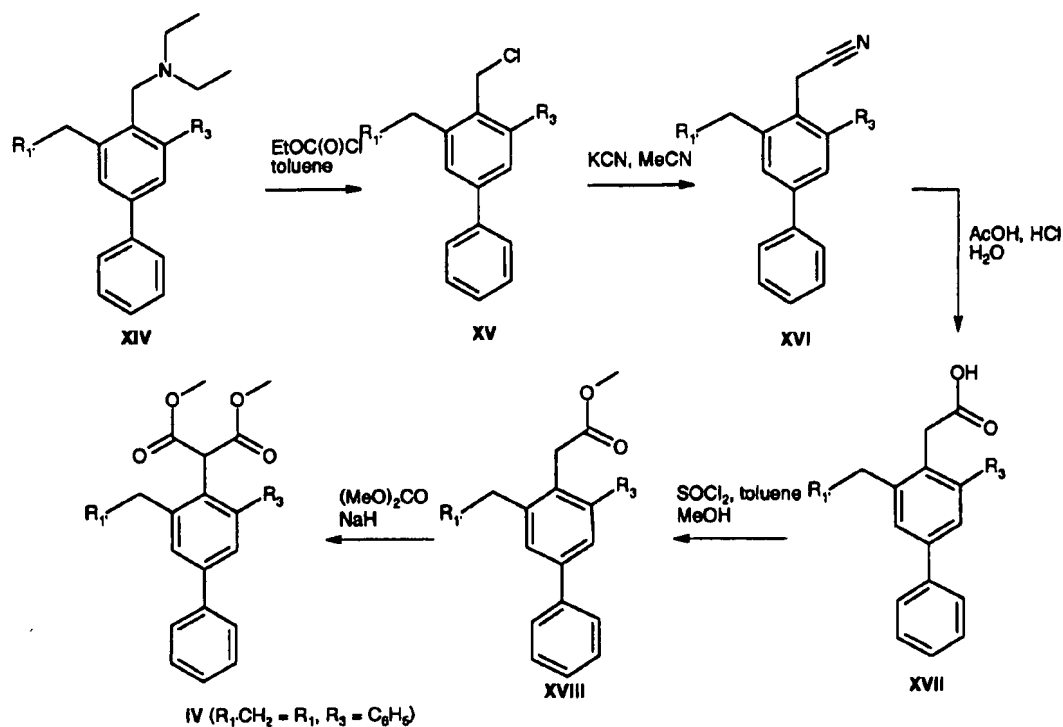
The palladium catalysts are used in an amount of from 0.001 to 50 mol %, preferably in an amount of from 0.1 to 15 mol %, based on the compound of formula VIII.

The reaction temperatures are selected in dependence upon the solvent used and, where applicable, the pressure. The reaction is preferably carried out at atmospheric pressure.

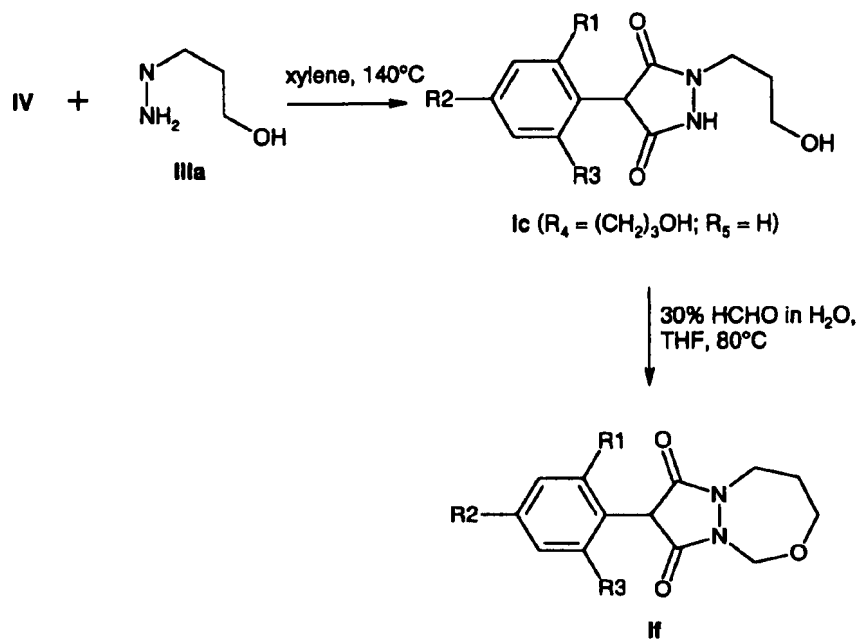
A further synthesis variant involves the ortho-functionalisation of a bi-aromatic compound: in that process, it is to be noted that any ortho-position CH_2 that is present must be protected against competitive metallisation before the introduction of the second ortho substituent (R_3).



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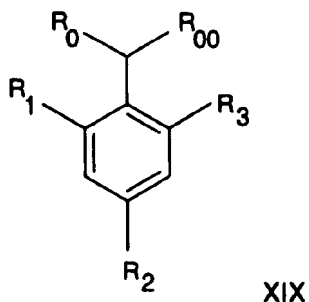


Compounds of the If type can be cyclised by amination with aldehydes (in this case with formaldehyde). Ic is accessible by the reaction of hydrazine alcohols, e.g. IIIa, with IV



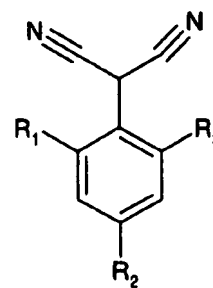
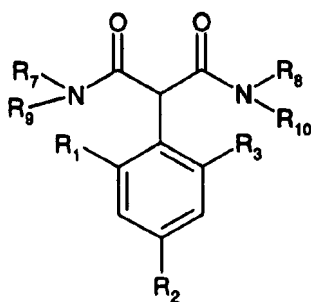
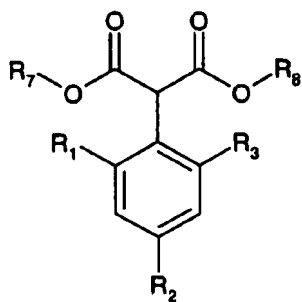
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The following intermediates of formula XIX, which are used in the above-mentioned syntheses and were developed specifically therefor, are novel and also form part of the present invention:



wherein R_0 is COOR_7 , COOR_8 , CONR_7R_9 , $\text{CONR}_8\text{R}_{10}$ or cyano, and R_{00} is hydrogen, COOR_7 , COOR_8 , CONR_7R_9 , $\text{CONR}_8\text{R}_{10}$ or cyano, and R_7 , R_8 , R_9 and R_{10} and R_1 , R_2 and R_3 are as defined above, but R_1 and R_3 are not simultaneously hydrogen.

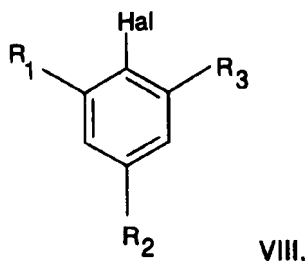
Preferred intermediates correspond to the formulae



wherein the substituents are as defined above.

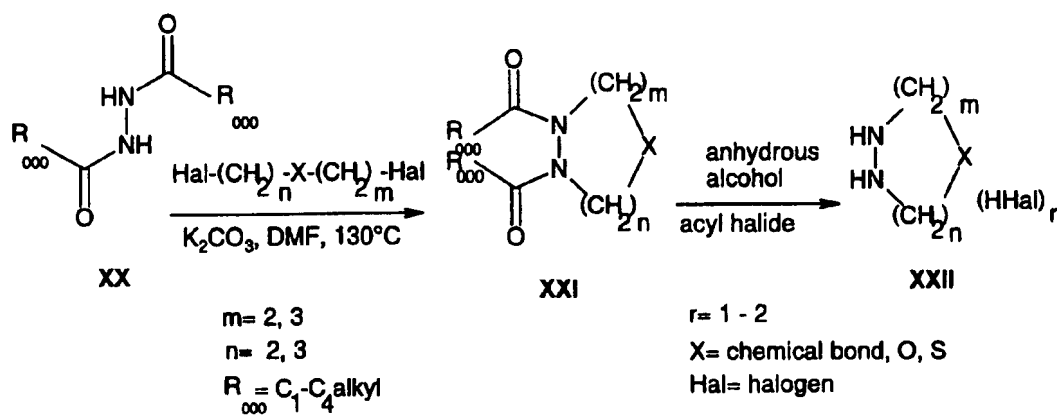
The following intermediates of formula VIII, which are used in the syntheses described above and were developed specifically therefor, are novel and also form part of the present invention:

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wherein R_1 , R_2 and R_3 are as defined above, and Hal is chlorine, bromine or iodine, Hal being other than iodine when R_1 and R_3 are methyl and R_2 is phenyl.

Hydrazine components required for the compounds according to the invention can be prepared according to a novel process. That process accordingly also forms part of the present invention. That process is characterised in that the last step is carried out in anhydrous alcohol directly with anhydrous hydrohalic acid or with hydrohalic acid prepared *in situ*, e.g. from the action of methanol on acetyl chloride.

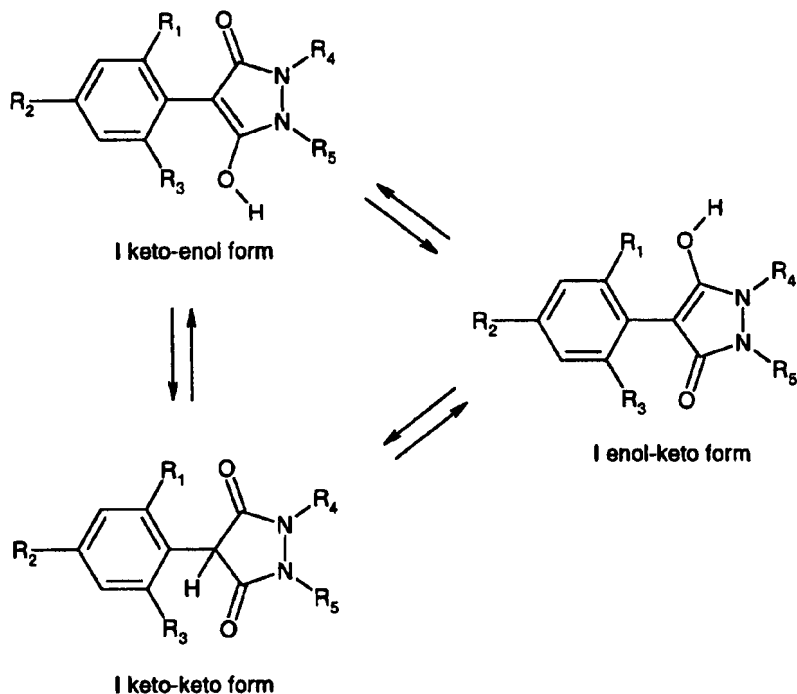


The hydrohalic acid can be produced by reacting anhydrous alcohol, such as methanol, with an acyl halide, such as acetyl halide, *in situ*, which is preferred. A preferred hydrohalic acid is hydrochloric acid.

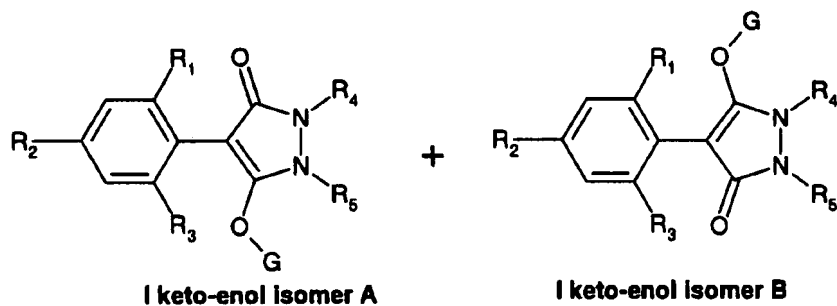
The substituent G can be introduced by the reaction of compounds of type I with organic or inorganic acids under water-removing conditions or in the presence of a coupling reagent. Acid chlorides and acid anhydrides are also very suitable for that purpose. It is to be noted that, in dependence on the type of substituents R_1 , R_3 , R_4 and $R_5 - R_{10}$, the compound of

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formula I may be in the form of a geometric and/or optical isomeric mixture or in the form of a tautomeric mixture. When $G = H$, for example, compound I may be present as the following three tautomers in equilibrium:



For that reason, when $R_4 \neq R_5$ the introduction of G can result in the formation of two additional geometric isomers:



The reactions to form compounds of formula I are advantageously carried out in aprotic, inert organic solvents. Such solvents are hydrocarbons, such as benzene, toluene, xylene or cyclohexane, chlorinated hydrocarbons, such as dichloromethane, trichloromethane, tetrachloromethane or chlorobenzene, ethers, such as diethyl ether, ethylene glycol dimethyl ether, diethylene glycol dimethyl ether, tetrahydrofuran or dioxane, nitriles, such as

acetonitrile or propionitrile, amides, such as N,N-dimethylformamide, diethylformamide or N-methylpyrrolidinone. The reaction temperatures are preferably from -20°C to +120°C. The reactions are generally slightly exothermic and can usually be carried out at room temperature. To reduce the reaction time or to initiate the reaction, the reaction mixture may also optionally be heated to boiling point for a short time. The reaction times can also be shortened by the addition of a few drops of base as reaction catalyst. Suitable bases are especially tertiary amines, such as trimethylamine, triethylamine, quinuclidine, 1,4-diazabicyclo[2.2.2]octane, 1,5-diazabicyclo[4.3.0]non-5-ene or 1,5-diazabicyclo[5.4.0]undec-7-ene. As bases it is also possible, however, to use inorganic bases, e.g. hydrides, such as sodium or calcium hydride, hydroxides, such as sodium or potassium hydroxide, carbonates, such as sodium and potassium carbonate, or hydrogen carbonates, such as potassium and sodium hydrogen carbonate. The compounds of formula I can be isolated in customary manner by concentration and/or evaporation of the solvent and can be purified by recrystallisation or trituration of the solid residue in solvents in which they are not readily soluble, such as ethers, aromatic hydrocarbons or chlorinated hydrocarbons.

For the use of the compounds of formula I according to the invention or compositions comprising them, there are suitable any of the methods of application customary in agriculture, such as preemergence application, postemergence application and seed dressing, as well as various methods and techniques, such as the controlled release of active ingredient. In that method the active ingredient is applied in solution to mineral granule carriers or polymerised granules (urea/formaldehyde) and dried. Where appropriate, it is also possible to apply a coating (coated granules) which allows the active ingredient to be released in metered amounts over a specific period.

The compounds of formula I can be used as herbicides in unmodified form, i.e. as obtained during synthesis, but are preferably formulated in customary manner together with the adjuvants conventionally employed in formulation technology, e.g. into emulsifiable concentrates, directly sprayable or dilutable solutions, dilute emulsions, wettable powders, soluble powders, dusts, granules and microcapsules. Such formulations are described, for example, in WO 97/34485 on pages 9 to 13. As with the nature of the compositions, the methods of application, such as spraying, atomising, dusting, wetting, scattering or pouring, are chosen in accordance with the intended objectives and the prevailing circumstances.

The formulations, i.e. the compositions, preparations or mixtures comprising the active ingredient of formula I or at least one active ingredient of formula I and generally one or more solid or liquid formulation adjuvants, are prepared in known manner, e.g. by intimately mixing and/or grinding the active ingredients with the formulation adjuvants, e.g. solvents or solid carriers. Surface-active compounds (surfactants) may additionally be used in the preparation of the formulations. Examples of solvents and solid carriers are given, for example, in WO 97/34485 on page 6.

Depending on the nature of the active ingredient of formula I to be formulated, suitable surface-active compounds are non-ionic, cationic and/or anionic surfactants and surfactant mixtures having good emulsifying, dispersing and wetting properties.

Examples of suitable anionic, non-ionic and cationic surfactants are listed, for example, in WO 97/34485 on pages 7 and 8. The surfactants customarily employed in formulation technology, which are described *inter alia* in "Mc Cutcheon's Detergents and Emulsifiers Annual" MC Publishing Corp., Ridgewood New Jersey, 1981, Stache, H., "Tensid-Taschenbuch", Carl Hanser Verlag, Munich/Vienna, 1981 and M. and J. Ash, "Encyclopedia of Surfactants", Vol I-III, Chemical Publishing Co., New York, 1980-81, are also suitable for the preparation of the herbicidal compositions according to the invention.

The herbicidal formulations usually comprise from 0.1 to 99 % by weight, especially from 0.1 to 95 % by weight, herbicide, from 1 to 99.9 % by weight, especially from 5 to 99.8 % by weight, of a solid or liquid formulation adjuvant and from 0 to 25 % by weight, especially from 0.1 to 25 % by weight, of a surfactant. Whereas commercial products are preferably formulated as concentrates, the end user will normally employ dilute formulations. The compositions may also comprise further ingredients such as stabilisers, e.g. vegetable oils and epoxidised vegetable oils (epoxidised coconut oil, rapeseed oil or soybean oil), antifoams, e.g. silicone oil, preservatives, viscosity regulators, binders, tackifiers, as well as fertilisers or other active ingredients.

The active ingredients of formula I are generally used on the plant or on the locus thereof at rates of application of from 0.001 to 4 kg/ha, especially from 0.005 to 2 kg/ha. The concentration required to achieve the desired effect can be determined by experiment. It is

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dependent upon the type of action, the stage of development of the crop plant and of the weed, and also upon the application (place, time, method) and, in dependence on those parameters, can vary within wide ranges.

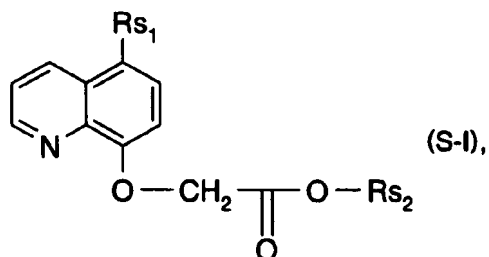
The compounds of formula I are distinguished by herbicidal and growth-inhibiting properties that make them suitable for use in crops of useful plants, especially in cereals, cotton, soybeans, sugar beet, sugar cane, plantation crops, rape, maize and rice, and for non-selective weed control. Crops are also to be understood as being those which have been rendered tolerant to herbicides or classes of herbicide by conventional methods of breeding or by genetic engineering techniques, e.g. IMI Maize, Poast Protected Maize, Liberty Link Maize, B.t./Liberty Link Maize, IMI/Liberty Link Maize, IMI/Liberty Link/B.t. Maize, Roundup Ready Maize and Roundup Ready/B.t. Maize. The weeds to be controlled may be either monocotyledonous or dicotyledonous weeds, for example *Stellaria*, *Nasturtium*, *Agrostis*, *Digitaria*, *Avena*, *Setaria*, *Sinapis*, *Lolium*, *Solanum*, *Echinochloa*, *Scirpus*, *Monochoria*, *Sagittaria*, *Bromus*, *Alopecurus*, *Sorghum halepense*, *Rottboellia*, *Cyperus*, *Abutilon*, *Sida*, *Xanthium*, *Amaranthus*, *Chenopodium*, *Ipomoea*, *Chrysanthemum*, *Galium*, *Viola* and *Veronica*.

The compound of formula I can be mixed advantageously with a number of further known herbicides, preferably selected from the groups of sulfonylureas, ureas, chloroacetanilides, chloroacetamides, diphenyl ethers, nitroanilines, oxadiazolones, pyrazoles, triazines, (hetero)aryloxypropionic acids, carbamates, thiocarbamates, thiatriazines, cyclohexanediones, imidazolinones, triazolopyrimidinesulfonamides, pyrimidinyloxy pyridinecarboxylic acids and pyrimidinyloxybenzoic acids. As a result, for example, a substantial widening of the weed spectrum and in many cases also an increase in selectivity towards the useful plants is obtained. The mixing partners of the compound of formula I may also optionally be present in the form of esters or salts, as mentioned, for example, in The Pesticide Manual, Eleventh Edition, 1997, BCPC.

The compound of formula I and mixtures thereof with the further herbicides mentioned above can also be used in combination with safeners. Preference is given to the following suitable safeners:

a compound of formula S-I

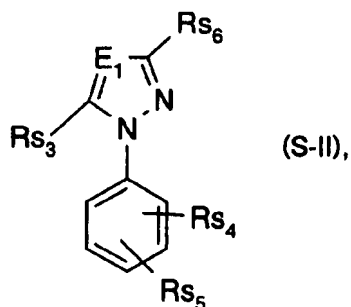
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wherein

Rs_1 is hydrogen or chlorine, and

Rs_2 is hydrogen, C_1 - C_8 alkyl or C_1 - C_8 alkyl substituted by C_1 - C_8 alkoxy or by C_3 - C_8 alkenyloxy, or a compound of formula S-II



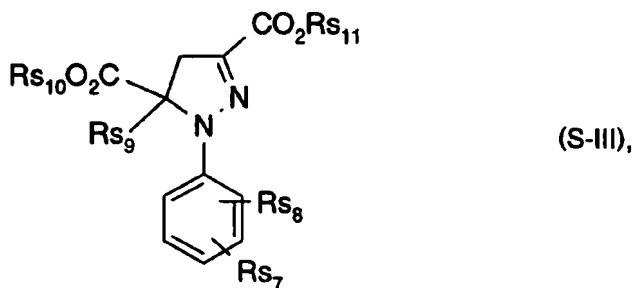
wherein E_1 is nitrogen or methine;

Rs_3 is $-CCl_3$, phenyl or halo-substituted phenyl;

Rs_4 and Rs_5 are each independently of the other hydrogen or halogen; and

Rs_6 is C_1 - C_4 alkyl;

or a compound of formula S-III

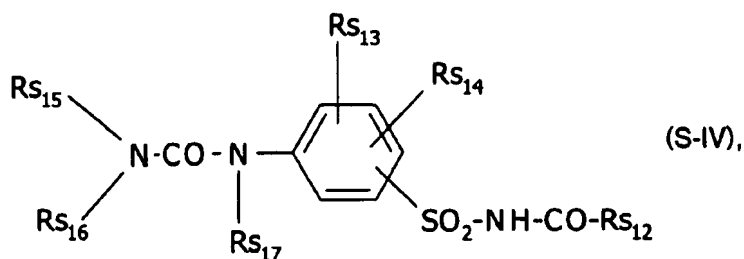


wherein Rs_7 and Rs_8 are each independently of the other hydrogen or halogen and

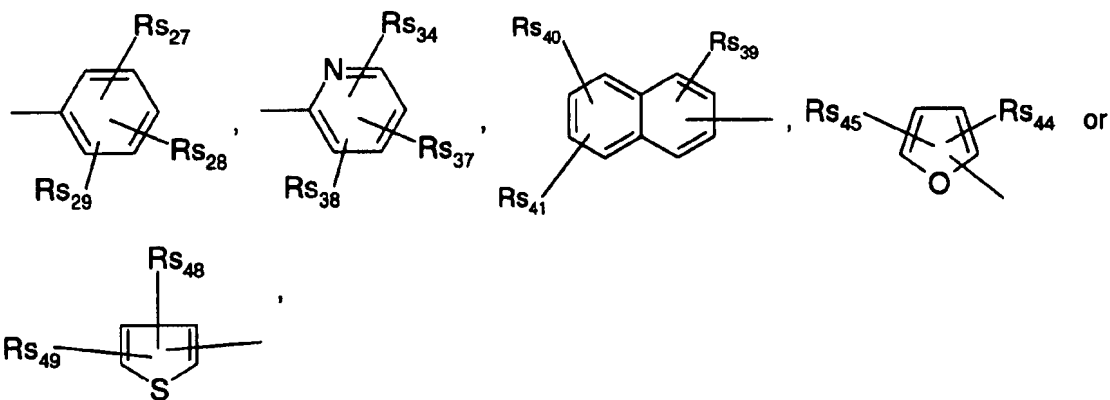
Rs_9 , Rs_{10} and Rs_{11} are each independently of the others C_1 - C_4 alkyl,

or a compound of formula S-IV

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wherein Rs_{12} is a group



Rs_{13} is hydrogen, halogen, cyano, trifluoromethyl, nitro, C_1 - C_4 alkyl, C_1 - C_4 alkoxy, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, $-COOH$, $-COO-C_1-C_4$ alkyl, $-CONRs_{18}Rs_{19}$, $-C(O)-C_1-C_4$ alkyl, $C(O)$ -phenyl, or phenyl substituted by halogen, C_1 - C_4 alkyl, methoxy, nitro or by trifluoromethyl, or $-SO_2NRs_{20}Rs_{21}$ or $-OSO_2-C_1-C_4$ alkyl;

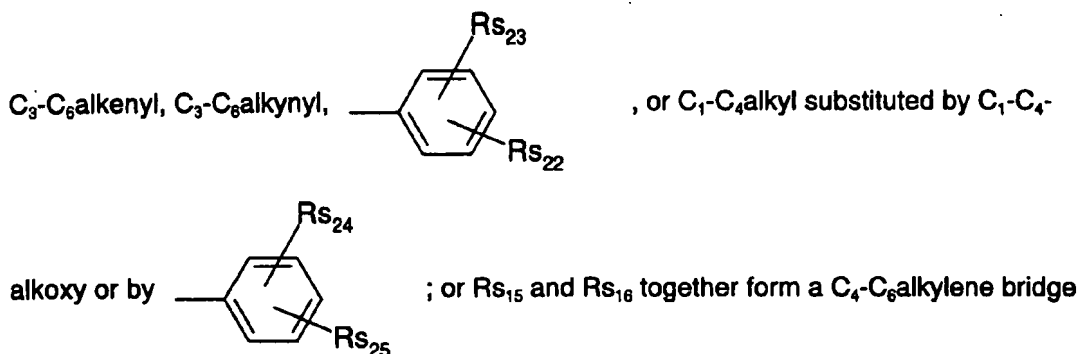
Rs_{18} , Rs_{19} , Rs_{20} and Rs_{21} are each independently of the others hydrogen or C_1 - C_4 alkyl, or Rs_{18} and Rs_{19} or Rs_{20} and Rs_{21} together form a C_4 - C_6 alkylene bridge which may be interrupted by oxygen, NH or by $-N(C_1-C_4$ alkyl)-;

Rs_{14} is hydrogen, halogen, C_1 - C_4 alkyl, trifluoromethyl, C_1 - C_6 alkoxy, C_1 - C_6 alkylthio, $-COOH$ or $-COO-C_1-C_4$ alkyl;

or Rs_{13} and Rs_{14} together form a C_3 - C_4 alkylene bridge which may be substituted by halogen or by C_1 - C_4 alkyl, or Rs_{13} and Rs_{14} together form a C_3 - C_4 alkenylene bridge which may be substituted by halogen or by C_1 - C_4 alkyl, or Rs_{13} and Rs_{14} together form a C_4 alkadienylene bridge which may be substituted by halogen or by C_1 - C_4 alkyl;

Rs_{15} and Rs_{16} are each independently of the other hydrogen, C_1 - C_6 alkyl, C_3 - C_8 cycloalkyl,

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which may be interrupted by oxygen, sulfur, SO, SO₂, NH or by -N(C_1-C_4 alkyl)-;

Rs_{22} , Rs_{23} , Rs_{24} and Rs_{25} are each independently of the others hydrogen, halogen, C_1-C_4 -alkyl, C_1-C_4 alkoxy, C_1-C_4 alkylthio, -COOR₂₆, trifluoromethyl, nitro or cyano, wherein Rs_{26} is in each case hydrogen, C_1-C_{10} alkyl, C_1-C_4 alkoxy- C_1-C_4 alkyl, C_1-C_4 alkylthio- C_1-C_4 alkyl, di- C_1-C_4 alkylamino- C_1-C_4 alkyl, halo- C_1-C_6 alkyl, C_2-C_8 alkenyl, halo- C_2-C_8 alkenyl, C_3-C_6 alkynyl, C_3-C_7 cycloalkyl, halo- C_3-C_7 cycloalkyl, C_1-C_8 alkylcarbonyl, allylcarbonyl, C_3-C_7 cycloalkylcarbonyl, benzoyl that is unsubstituted or substituted on the phenyl ring identically or differently up to three times by halogen, C_1-C_4 alkyl, halo- C_1-C_4 alkyl, halo- C_1-C_4 alkoxy or C_1-C_4 alkoxy; or furyl or thienyl; or C_1-C_4 alkyl substituted by phenyl, halophenyl, C_1-C_4 -alkylphenyl, C_1-C_4 alkoxyphenyl, halo- C_1-C_4 alkylphenyl, halo- C_1-C_4 alkoxyphenyl, C_1-C_6 -alkoxycarbonyl, C_1-C_4 alkoxy- C_1-C_6 alkoxycarbonyl, C_3-C_6 alkenyloxycarbonyl, C_3-C_6 alkynyloxycarbonyl, C_1-C_6 alkylthiocarbonyl, C_3-C_6 alkenylthiocarbonyl, C_3-C_6 alkynylthiocarbonyl, carbamoyl, mono- C_1-C_4 alkylaminocarbonyl or by di- C_1-C_4 alkylaminocarbonyl; or phenylaminocarbonyl which may itself be substituted on the phenyl identically or differently up to three times by halogen, C_1-C_4 alkyl, halo- C_1-C_4 alkyl, halo- C_1-C_4 alkoxy or C_1-C_4 alkoxy or once by cyano or nitro; or dioxolan-2-yl which may itself be substituted by one or two C_1-C_4 alkyl groups, or dioxan-2-yl which may itself be substituted by one or two C_1-C_4 alkyl groups; or C_1-C_4 alkyl substituted by cyano, nitro, carboxyl or by C_1-C_6 -alkylthio- C_1-C_6 alkoxycarbonyl; Rs_{17} is hydrogen or C_1-C_4 alkyl;

Rs_{27} is hydrogen, halogen, nitro, C_1-C_4 alkyl or methoxy;

Rs_{28} is hydrogen, halogen, C_1-C_4 alkyl, trifluoromethyl, C_1-C_6 alkoxy, C_1-C_6 alkylthio, -COOH or -COO- C_1-C_4 alkyl;

Rs_{29} is hydrogen, halogen, cyano, nitro, C_1-C_4 alkyl, C_1-C_4 haloalkyl, C_1-C_4 alkylthio, C_1-C_4 -alkylsulfinyl, C_1-C_4 alkylsulfonyl, -COOH, -COO- C_1-C_4 alkyl, -CONR₃₀R₃₁, C(O)-phenyl, or phenyl substituted by halogen, C_1-C_4 alkyl, methoxy, nitro or by trifluoromethyl, or

-SO₂NRs₃₂Rs₃₃, -OSO₂-C₁-C₄alkyl, C₁-C₆alkoxy, or C₁-C₆alkoxy substituted by C₁-C₄alkoxy or by halogen; or C₃-C₆alkenyloxy or C₃-C₆alkenyloxy substituted by halogen; or C₃-C₆alkynyl-oxy; wherein Rs₃₀ and Rs₃₁ are each independently of the other hydrogen or C₁-C₄alkyl; or Rs₃₀ and Rs₃₁ together form a C₄-C₆alkylene bridge which may be interrupted by oxygen, NH or by -N(C₁-C₄alkyl)-, and Rs₃₂ and Rs₃₃ are each independently of the other hydrogen or C₁-C₄alkyl, or Rs₃₂ and Rs₃₃ together form a C₄-C₆alkylene bridge which may be interrupted by oxygen, NH or by -N(C₁-C₄alkyl)-;

Rs₃₄ is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOH, -COO-C₁-C₄alkyl or CONRs₃₅Rs₃₆, wherein Rs₃₅ and Rs₃₆ are each independently of the other hydrogen or C₁-C₄alkyl, or Rs₃₅ and Rs₃₆ together form a C₄-C₆alkylene bridge which may be interrupted by oxygen, NH or by -N(C₁-C₄alkyl)-;

Rs₃₇ is hydrogen, halogen, C₁-C₄alkyl, -COOH, -COO-C₁-C₄alkyl, trifluoromethyl or methoxy, or Rs₃₄ and Rs₃₇ together form a C₃-C₄alkylene bridge;

Rs₃₈ is hydrogen, halogen or C₁-C₄alkyl;

Rs₃₉ is hydrogen, halogen, C₁-C₄alkyl, -COOH, -COO-C₁-C₄alkyl, trifluoromethyl or methoxy;

Rs₄₀ is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOH, -COO-C₁-C₄alkyl or CONRs₄₂Rs₄₃;

Rs₄₁ is hydrogen, halogen or C₁-C₄alkyl; or Rs₄₀ and Rs₄₁ together form a C₃-C₄alkylene bridge;

Rs₄₂ and Rs₄₃ are each independently of the other hydrogen or C₁-C₄alkyl, or Rs₄₂ and Rs₄₃ together form a C₄-C₆alkylene bridge which may be interrupted by oxygen, NH or by -N(C₁-C₄alkyl)-;

Rs₄₄ is hydrogen, halogen, C₁-C₄alkyl, -COOH, -COO-C₁-C₄alkyl, trifluoromethyl or methoxy;

Rs₄₅ is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOH, -COO-C₁-C₄alkyl or CONRs₄₆Rs₄₇;

Rs₄₆ and Rs₄₇ are each independently of the other hydrogen or C₁-C₄alkyl, or Rs₄₆ and Rs₄₇ together form a C₄-C₆alkylene bridge which may be interrupted by oxygen, NH or by -N(C₁-C₄alkyl)-;

Rs₄₈ is hydrogen, halogen, C₁-C₄alkyl, -COOH, -COO-C₁-C₄alkyl, trifluoromethyl or methoxy;

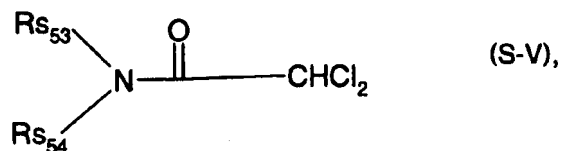
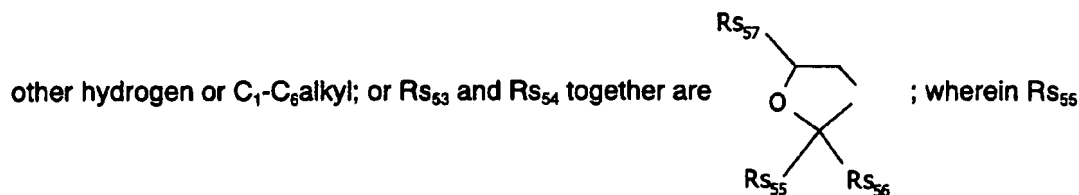
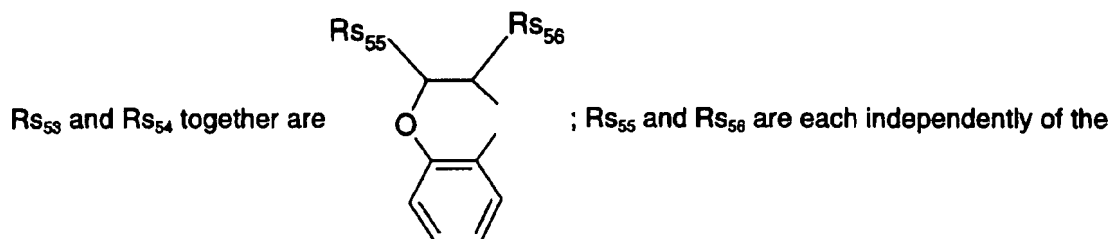
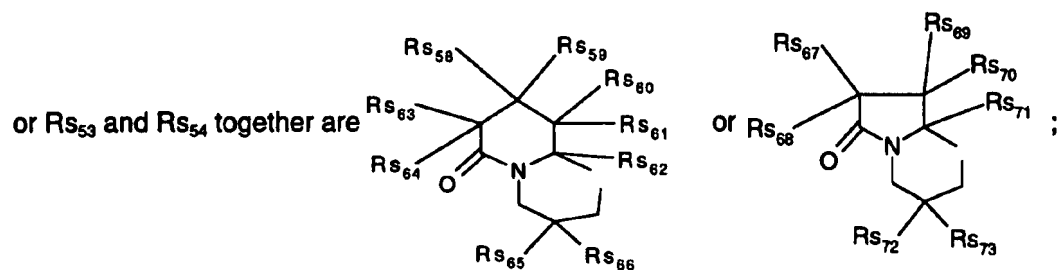
Rs₄₉ is hydrogen, halogen, nitro, C₁-C₄alkyl, C₁-C₄alkoxy, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, -COOH, -COO-C₁-C₄alkyl or CONRs₅₀Rs₅₁;

Rs₅₁ and Rs₅₂ are each independently of the other hydrogen or C₁-C₄alkyl, or Rs₅₁ and Rs₅₂ together form a C₄-C₆alkylene bridge which may be interrupted by oxygen, NH or by

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-N(C₁-C₄alkyl)-;

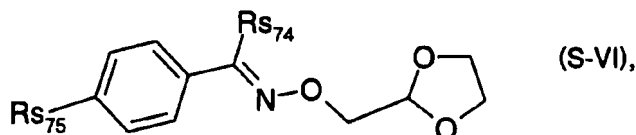
or a compound of formula S-V

wherein Rs₅₃ and Rs₅₄ are each independently of the other C₁-C₆alkyl or C₂-C₆alkenyl; orand Rs₅₆ are each independently of the other C₁-C₄alkyl, or Rs₅₅ and Rs₅₆ together are -(CH₂)₅;

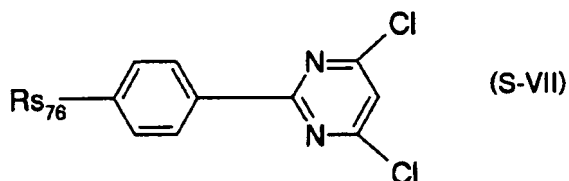
wherein

Rs₅₈, Rs₅₉, Rs₆₀, Rs₆₁, Rs₆₂, Rs₆₃, Rs₆₄, Rs₆₅, Rs₆₆, Rs₆₇, Rs₆₈, Rs₆₉, Rs₇₀, Rs₇₁, Rs₇₂ and Rs₇₃ are each independently of the others hydrogen or C₁-C₄alkyl; or a compound of formula S-VI

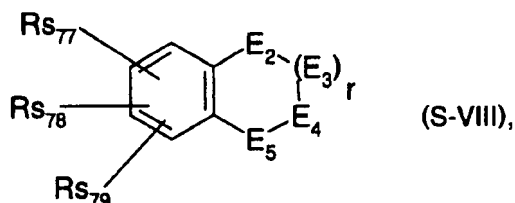
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wherein Rs_{75} is hydrogen or chlorine and Rs_{74} is cyano or trifluoromethyl,
or a compound of formula S-VII



wherein Rs_{76} is hydrogen or methyl,
or of formula S-VIII



wherein

r is 0 or 1;

Rs_{77} is hydrogen or C_1 - C_4 alkyl which may be substituted by C_1 - C_4 alkoxy, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfinyl, C_1 - C_4 alkylsulfonyl, C_1 - C_4 haloalkyl, C_1 - C_4 haloalkoxy, C_1 - C_4 haloalkylthio, C_1 - C_4 haloalkylsulfinyl, C_1 - C_4 haloalkylsulfonyl, nitro, cyano, -COOH, COO- C_1 - C_4 alkyl, -NR s_{80} Rs $_{81}$, -SO $_2$ NR s_{82} Rs $_{83}$ or by -CONR s_{84} Rs $_{85}$;

Rs_{78} is hydrogen, halogen, C_1 - C_4 alkyl, trifluoromethyl, C_1 - C_4 alkoxy or C_1 - C_4 haloalkoxy;

Rs_{79} is hydrogen, halogen or C_1 - C_4 alkyl;

Rs_{80} is hydrogen, C_1 - C_4 alkyl or C_1 - C_4 alkylcarbonyl;

Rs_{81} is hydrogen or C_1 - C_4 alkyl; or

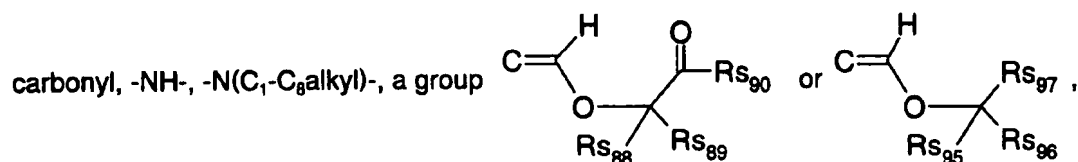
Rs_{80} and Rs_{81} together form a C_4 - or C_5 -alkylene group;

Rs_{82} , Rs_{83} , Rs_{84} and Rs_{85} are each independently of the others hydrogen or C_1 - C_4 alkyl; or

Rs_{82} together with Rs_{83} , or Rs_{84} together with Rs_{85} , are each independently of the other C_4 - or C_5 -alkylene, it being possible for one carbon atom to be replaced by oxygen or sulfur, or for one or two carbon atoms to be replaced by -NH- or by -N(C_1 - C_4 alkyl)-;

E_2 , E_3 , E_4 and E_5 are each independently of the others oxygen, sulfur, C(Rs $_{86}$)Rs $_{87}$,

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Rs₈₆ and Rs₈₇ are each independently of the other hydrogen or C₁-C₈alkyl; or

Rs₈₆ and Rs₈₇ together are C₂-C₈alkylene;

Rs₈₈ and Rs₈₉ are each independently of the other hydrogen or C₁-C₈alkyl; or

Rs₈₈ and Rs₈₉ together form a C₂-C₈alkylene group;

Rs₉₀ is Rs₉₁-O-, Rs₉₂-S- or -NR₉₃Rs₉₄;

Rs₉₁ and Rs₉₂ are each independently of the other hydrogen, C₁-C₈alkyl, C₁-C₈haloalkyl, C₁-C₄alkoxy-C₁-C₈alkyl, C₃-C₈alkenyloxy-C₁-C₈alkyl or phenyl-C₁-C₈alkyl, it being possible for the phenyl ring to be substituted by halogen, C₁-C₄alkyl, trifluoromethyl, methoxy, methylthio, methylsulfinyl or by methylsulfonyl, or are C₃-C₈alkenyl, C₃-C₈haloalkenyl, phenyl-C₃-C₈alkenyl, C₃-C₈alkynyl, phenyl-C₃-C₈alkynyl, oxetanyl, furyl or tetrahydrofuryl;

Rs₉₃ is hydrogen, C₁-C₈alkyl, phenyl, phenyl-C₁-C₈alkyl, it being possible for the phenyl rings to be substituted by fluorine, chlorine, bromine, nitro, cyano, -OCH₃, C₁-C₄alkyl or by CH₃SO₂-, or is C₁-C₄alkoxy-C₁-C₈alkyl, C₃-C₈alkenyl or C₃-C₈alkynyl;

Rs₉₄ is hydrogen, C₁-C₈alkyl, C₃-C₈alkenyl or C₃-C₈alkynyl; or

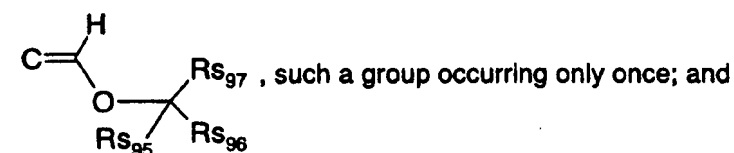
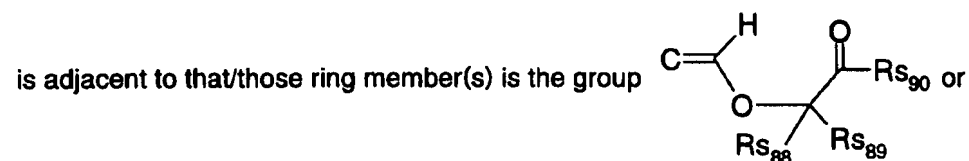
Rs₉₃ and Rs₉₄ together are C₄- or C₅-alkylene, it being possible for one carbon atom to be replaced by oxygen or sulfur, or for one or two carbon atoms to be replaced by -NH- or by -N(C₁-C₄alkyl)-;

Rs₉₅ and Rs₉₆ are each independently of the other hydrogen or C₁-C₈alkyl; or

Rs₉₅ and Rs₉₆ together form a C₂-C₈alkylene group; and

Rs₉₇ is C₂-C₄alkenyl or C₂-C₄alkynyl; with the provisos that

a) at least one of the ring members E₂, E₃, E₄ and E₅ is carbonyl, and one ring member that

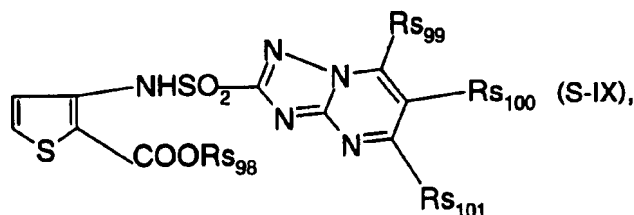


b) two adjacent ring members E₂ and E₃, E₃ and E₄, and E₄ and E₅ cannot simultaneously be

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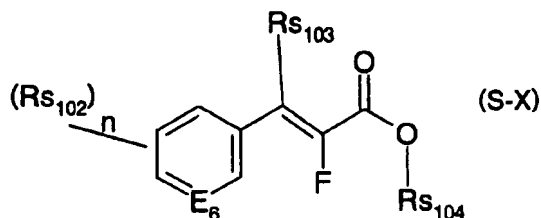
oxygen;

or a compound of formula S-IX



wherein Rs_{98} is hydrogen, C_1 - C_6 alkyl, C_3 - C_6 cycloalkyl, C_3 - C_6 alkenyl or C_3 - C_6 alkynyl; and Rs_{99} , Rs_{100} and Rs_{101} are each independently of the others hydrogen, C_1 - C_6 alkyl, C_3 - C_6 cycloalkyl or C_1 - C_6 alkoxy, with the proviso that one of the substituents Rs_{99} , Rs_{100} and Rs_{101} is other than hydrogen;

or a compound of formula S-X



wherein E_6 is nitrogen or methine, n is 0, 1, 2 or 3 when E_6 is nitrogen and 0, 1, 2, 3 or 4 when E_6 is methine, Rs_{102} is halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 haloalkoxy, nitro, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkoxycarbonyl, phenyl or phenoxy, or phenyl or phenoxy each substituted by C_1 - C_3 alkyl, C_1 - C_3 haloalkyl, C_1 - C_3 alkoxy, C_1 - C_3 haloalkoxy, halogen, cyano or by nitro;

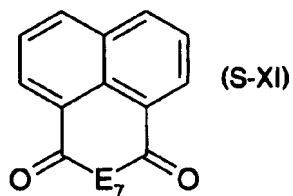
Rs_{103} is hydrogen or C_1 - C_4 alkyl;

Rs_{104} is hydrogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_2 - C_6 alkenyl, C_2 - C_6 alkynyl, C_1 - C_4 haloalkyl, C_2 - C_6 haloalkenyl, C_2 - C_6 haloalkynyl, C_1 - C_4 alkylthio- C_1 - C_4 alkyl, C_1 - C_4 alkylsulfonyl- C_1 - C_4 alkyl, C_1 - C_4 alkoxy- C_1 - C_4 alkyl, C_1 - C_4 alkenyloxy- C_1 - C_4 alkyl or C_1 - C_4 alkynyloxy- C_1 - C_4 alkyl;

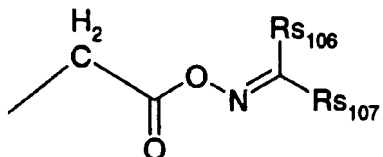
or a compound of formula S-XI

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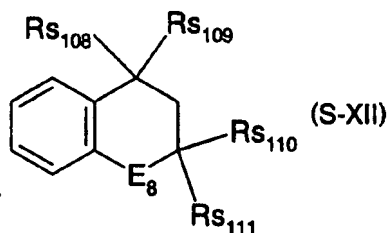


wherein E_7 is oxygen or N- Rs_{105} and Rs_{105} is a group of formula



wherein Rs_{106} and Rs_{107} are each independently of the other cyano, hydrogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_2 - C_6 alkenyl, aryl, phenyl or heteroaryl, or phenyl, aryl or heteroaryl each substituted by C_1 - C_3 alkyl, C_1 - C_3 haloalkyl, C_1 - C_3 alkoxy, C_1 - C_3 haloalkoxy, halogen, cyano or by nitro;

or a compound of formula S-XII

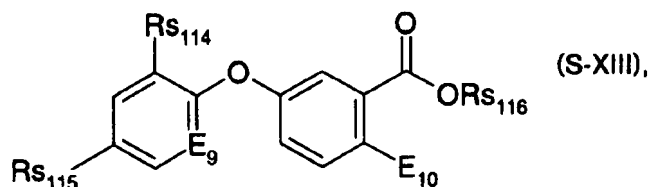


wherein E_8 is oxygen, sulfur, sulfinyl, sulfonyl or methine,

Rs_{108} and Rs_{109} are each independently of the other CH_2COORs_{112} or $COORs_{113}$ or together are a group of formula $-(CH_2)C(O)-O-C(O)-(CH_2)-$, and Rs_{112} and Rs_{113} are each independently of the other hydrogen, C_1 - C_4 alkyl, C_2 - C_4 alkenyl, C_2 - C_6 alkynyl, C_3 - C_6 cycloalkyl, C_1 - C_4 haloalkyl, or a metal cation or ammonium cation; and

Rs_{110} and Rs_{111} are each independently of the other hydrogen, halogen or C_1 - C_4 alkyl; or a compound of formula S-XIII

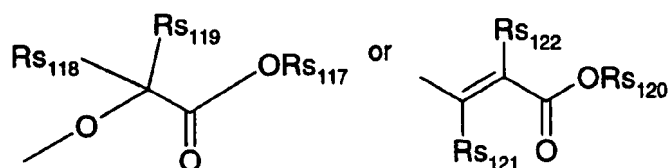
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wherein Rs_{114} and Rs_{115} are each independently of the other hydrogen, halogen or C_1 - C_4 -haloalkyl, Rs_{116} is hydrogen, C_1 - C_4 alkyl, C_3 - C_4 alkenyl, C_3 - C_4 alkynyl, C_1 - C_4 haloalkyl, C_3 - C_6 -cycloalkyl, a metal cation or an ammonium cation;

E_9 is nitrogen, methine, C-F or C-Cl and

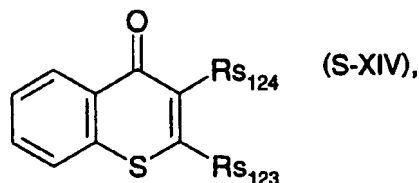
E_{10} is a group of formula



, wherein Rs_{118} , Rs_{119} , Rs_{121} and

Rs_{122} are each independently of the others hydrogen or C_1 - C_4 alkyl;

Rs_{117} and Rs_{120} are each independently of the other hydrogen, C_1 - C_4 alkyl, C_3 - C_4 alkenyl, C_3 - C_4 alkynyl, C_1 - C_4 haloalkyl, C_3 - C_6 cycloalkyl, a metal cation or an ammonium cation; or a compound of formula S-XIV



wherein Rs_{123} is hydrogen, cyano, halogen, C_1 - C_4 alkyl, C_3 - C_6 cycloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 -alkoxycarbonyl, C_1 - C_4 alkylthiocarbonyl, $-NH-Rs_{125}-C(O)NH-Rs_{126}$, aryl or heteroaryl, or aryl or heteroaryl each substituted by C_1 - C_3 alkyl, C_1 - C_3 haloalkyl, C_1 - C_3 alkoxy, C_1 - C_3 haloalkoxy, halogen, cyano or by nitro;

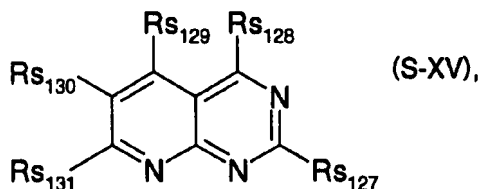
Rs_{124} is hydrogen, cyano, nitro, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, C_1 - C_4 -thioalkyl; and

Rs_{125} and Rs_{126} are each independently of the other C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_3 - C_4 alkenyl, C_3 - C_4 alkynyl, C_3 - C_6 cycloalkyl, C_1 - C_4 alkylcarbonyl, C_1 - C_4 alkylsulfonyl, aryl or heteroaryl, or aryl or heteroaryl each substituted by C_1 - C_3 alkyl, C_1 - C_3 haloalkyl, C_1 - C_3 alkoxy, C_1 - C_3 halo-

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alkoxy, halogen, cyano or by nitro;

or a compound of formula S-XV

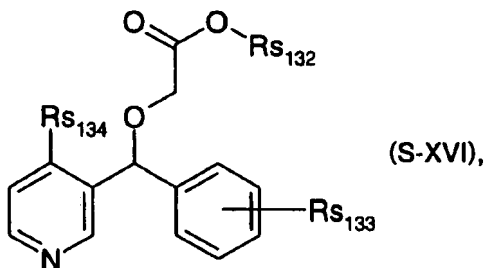


wherein Rs_{127} and Rs_{128} are each independently of the other hydrogen, C_1 - C_4 alkyl, C_1 - C_4 -haloalkyl, C_1 - C_4 alkoxy, mono- C_1 - C_8 - or di- C_1 - C_8 -alkylamino, C_3 - C_6 cycloalkyl, C_1 - C_4 thioalkyl, phenyl or heteroaryl;

Rs_{129} is hydrogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, mono- C_1 - C_8 - or di- C_1 - C_8 -alkylamino, C_3 - C_6 cycloalkyl, C_1 - C_4 thioalkyl, phenyl, heteroaryl, OH, NH_2 , halogen, di- C_1 - C_4 -aminoalkyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl or C_1 - C_4 alkoxycarbonyl;

Rs_{130} is hydrogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, mono- C_1 - C_8 - or di- C_1 - C_8 -alkylamino, C_3 - C_6 cycloalkyl, C_1 - C_4 thioalkyl, phenyl, heteroaryl, cyano, nitro, carboxyl, C_1 - C_4 -alkoxycarbonyl, di- C_1 - C_4 -aminoalkyl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, SO_2 -OH, i- C_1 - C_4 -aminoalkylsulfonyl or C_1 - C_4 alkoxysulfonyl;

Rs_{131} is hydrogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl, C_1 - C_4 alkoxy, mono- C_1 - C_8 - or di- C_1 - C_8 -alkylamino, C_3 - C_6 cycloalkyl, C_1 - C_4 thioalkyl, phenyl, heteroaryl, OH, NH_2 , halogen, di- C_1 - C_4 -aminoalkyl, pyrrolidin-1-yl, piperid-1-yl, morpholin-1-yl, C_1 - C_4 alkylthio, C_1 - C_4 alkylsulfonyl, C_1 - C_4 alkoxycarbonyl, phenoxy, naphthoxy, phenylamino, benzoyloxy or phenylsulfonyloxy; or a compound of formula S-XVI



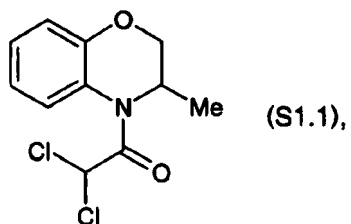
wherein Rs_{132} is hydrogen, C_4 alkyl, C_1 - C_4 haloalkyl, C_2 - C_4 alkenyl, C_2 - C_4 alkynyl or C_1 - C_4 -alkoxy- C_1 - C_4 alkyl;

Rs_{133} is hydrogen, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or C_1 - C_4 alkoxy and Rs_{134} is hydrogen, halogen, C_1 - C_4 alkyl, C_1 - C_4 haloalkyl or C_1 - C_4 alkoxy; with the proviso that Rs_{133} and Rs_{134} are

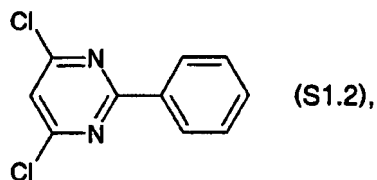
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not simultaneously hydrogen.

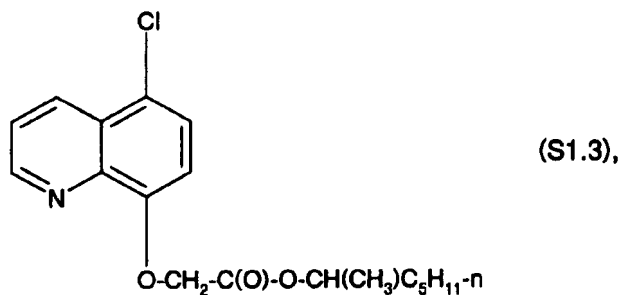
Especially preferred safeners for the composition according to the invention are selected from the group of compounds of formula S1.1



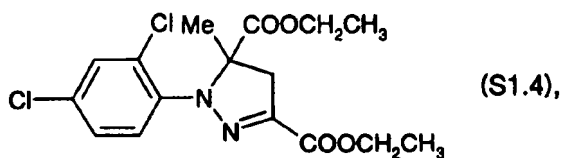
and of the compound of formula S1.2



and of the compound of formula SS1.3

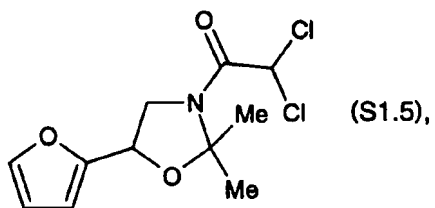


and of the compound of formula S1.4

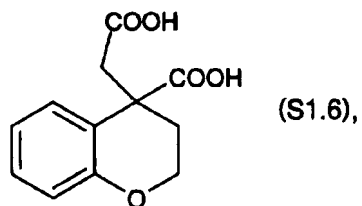


and of the compound of formula S1.5

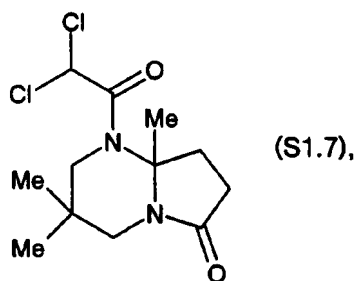
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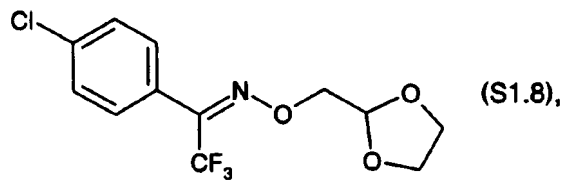
and of the compound of formula S1.6



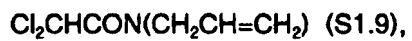
and of the compound of formula S1.7



and of the compound of formula S1.8

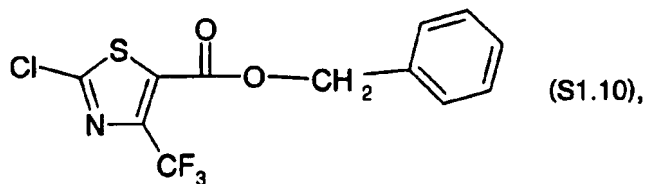


and of formula S1.9

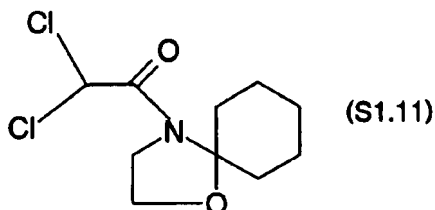


and of formula S1.10

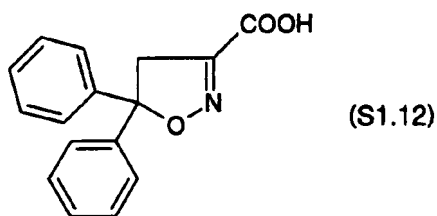
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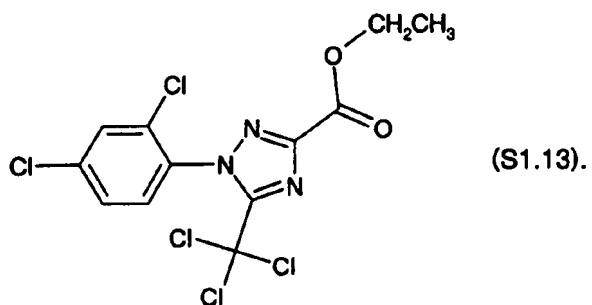
and of formula S1.11



and of formula S1.12



and of formula S1.13



The compounds of formulae S1.1 to S1.13 are known and are described, for example in The Pesticide Manual, eleventh ed., British Crop Protection Council, 1997 under entry numbers 61 (formula S1.1, benoxacor), 304 (formula S1.2, fenclorim), 154 (formula S1.3, cloquintocet), 462 (formula S1.4, mefenpyr-diethyl), 377 (formula S1.5, furilazole), 363 (formula S1.8, fluxofenim), 213 (formula S1.9, dichlormid) and 350 (formula S1.10, flurazole). The compound of formula S1.11 is known by the name MON 4660 (Monsanto)

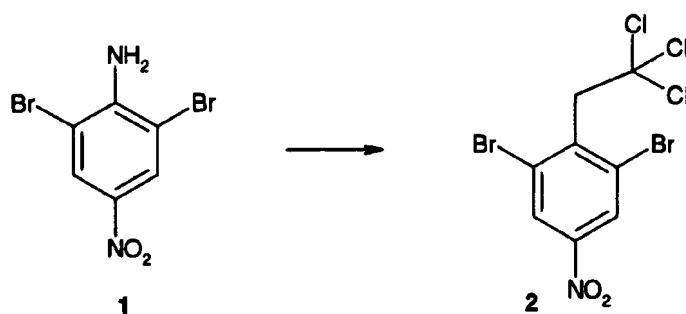
and is described, for example, in EP-A-0 436 483.

The compound of formula S1.6 (AC 304415) is described, for example, in EP-A-0 613 618, and the compound of formula S1.7 is described in DE-A-2948535. The compound of formula S1.12 is described in DE-A-4331448, and the compound of formula S1.13 is described in DE-A-3525205.

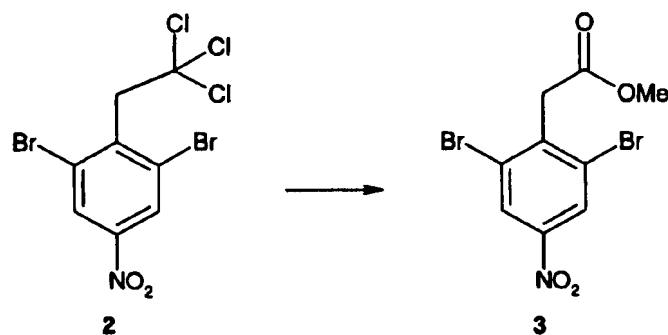
The following Examples illustrate the invention further but do not limit it.

Preparation Examples:

Example P1: Preparation of 3-hydroxy-4-aryl-5-oxo-pyrazolines

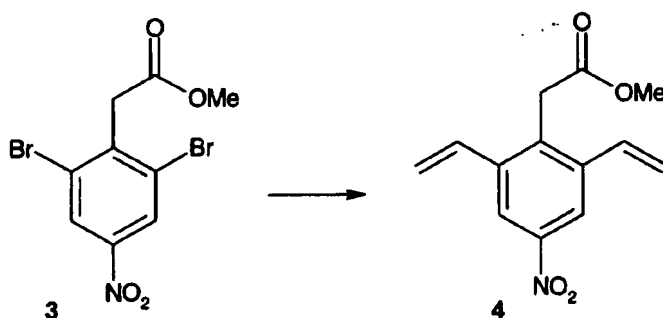


Vinylidene chloride (84 ml, 1.05 M), tert-butyl nitrite (12.5 ml, 0.105 M) and copper(II) chloride were placed in acetonitrile (70 ml). At 10°C, 2,6-dibromo-4-nitroaniline **1** was added in portions thereto. The reaction mixture was then stirred at room temperature for 16 hours and filtered, and the filtrate was washed with tert-butyl methyl ether. The organic phase was washed with 150 ml of 10% HCl and 2 x 150 ml of water, and dried over Na₂SO₄. The organic phase was concentrated and chromatographed over silica gel. 13.2 g of **2** were obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 4.69 (s, 2H).



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2 (10.9 g, 26.4 mmol) was suspended in methanol and an NaOMe solution (30% in methanol, 27.8 ml) was added at room temperature. The reaction mixture was then refluxed for 1 hour, cooled, rendered acidic with H₂SO₄ (3.7 ml) and refluxed for a further 0.5 hour. The reaction mixture was cooled, diluted with water and extracted with CH₂Cl₂ (3 x 100 ml). The combined organic phases were dried using Na₂SO₄, filtered and concentrated. **3** (9.1 g) was obtained in the form of a crude product. ¹H-NMR (300 MHz, CDCl₃): δ = 8.4 (s, 2H); 4.2 (s, 2H); 3.7 (s, 3H).

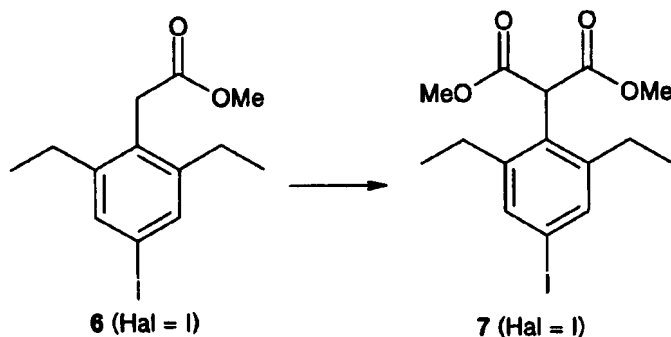


3 (9.1, 25.7 mmol) and vinyl tributyl tin (18 ml, 61.7 mmol) were placed in toluene. The reaction mixture was degassed for 0.33 hour by the introduction of argon. Tetrakis-triphenylphosphine-Pd(0) (1.46 g, 1.28 mmol) was then added thereto. The reaction mixture was heated at 100°C for 16 hours and cooled, and an NaOH solution (1N, 100 ml) was added. The two-phase mixture was stirred rapidly for 0.5 hour. The organic phase was separated off and washed with water and brine, dried over Na₂SO₄, concentrated and chromatographed over silica gel (EtOAc:hexane (1:10)). **4** (4.13 g) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 8.2 (s, 2H); 6.9 (dd, 1H); 5.8 (d, 1H); 5.5 (d, 1H); 3.8 (s, 2H); 3.7 (s, 3H).

4 (4 g, 16.2 mmol) was dissolved in MeOH (100 ml), and Pd/C (5%, 2 g) was added thereto. The reaction mixture was stirred rapidly for 2.5 hours under a hydrogen atmosphere (normal pressure). The reaction mixture was filtered and concentrated. **5** (3.25 g) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 6.4 (s, 2H); 3.7 (s, 3H); 3.6 (s, 2H); 2.6 (q, 4H); 1.2 (t, 6H)

Tert-butyl nitrite (5.96 ml, 50 mmol) and methylene iodide (4.15 ml, 50 mmol) were placed in acetonitrile (30 ml) and, at 0°C, **5** (5.52 g, 25 mmol) dissolved in acetonitrile (20 ml), was added thereto. The reaction mixture was illuminated for 1 hour (200 W), during which the temperature rose to 70°C. The reaction mixture was cooled, water was added, and

extraction was carried out with ethyl acetate (2 x 20 ml). The combined organic phases were washed with 1N HCl and brine, dried over Na_2SO_4 , filtered, concentrated and chromatographed over silica gel (ethyl acetate:hex = 1:5). **6** (Hal = I) (1.6 g) was obtained. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.4 (s, 2H); 3.7 (2s, 5H); 2.6 (q, 4H); 1.2 (t, 6H).



Diisopropylamine (565 mg, 5.59 mmol) was placed in THF (15 ml) and, at -30°C , *n*-BuLi (2M in cyclohexane, 2.67 ml, 5.34 mmol) was added. The reaction mixture was stirred at 0°C for 0.25 hour and cooled to -78°C . **6** (Hal = I) was dissolved in THF (15 ml) and added dropwise to the reaction mixture. After stirring for 0.5 hour at -78°C , cyanoformic acid methyl ester was added, and the reaction mixture was slowly heated to 0°C . The reaction mixture was added to NH_4Cl (aq) (100 ml) and extracted with ethyl acetate (2 x 70 ml). The combined organic phases were washed with H_2O (3 x 70 ml) and brine (70 ml), dried over Na_2SO_4 , filtered and concentrated. The crude product was chromatographed over silica gel.

7 (Hal = I, 1.125 g) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 7.4 (s, 2H); 5.0 (s, 1H); 3.8 (s, 6H); 2.6 (m, 4H) 1.2 (m, 6H).

Tert-butyl nitrite (355 ml, 3 mol) and copper(II) chloride (337.4 g, 2.5 mol) were placed in acetonitrile (1200 ml). First, at < 30°C, vinylidene chloride (2385 ml, 29.9 mol) was added dropwise and then, at 10°C, a solution of 4-bromo-2,6-dimethylaniline **8** (398 g, 2 mol) in acetonitrile (2000 ml) was added dropwise. The reaction mixture was then stirred at room temperature for 16 hours, stirred into 20% HCl (9000 ml) and extracted with tert-butyl methyl ether (3 x 3000 ml). The organic phase was washed with 20% HCl and water and dried over Na₂SO₄. The organic phase was concentrated. 470 g of **9** were obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 4.13 (s, 2H).

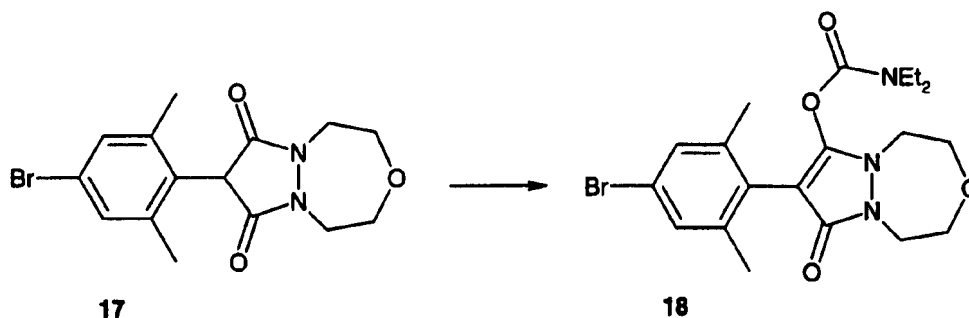
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9 (257 g, 0.813 mol) was dissolved in methanol (400 ml) and, at < 30°C, NaOMe (30% in methanol, 640 ml, 3.53 mol) was added. The reaction mixture was stirred at reflux for 15 hours. The reaction mixture was cooled to room temperature, and concentrated sulfuric acid (95 ml, 1.75 mol) was added. The reaction mixture was stirred at reflux for 10 hours, concentrated and stirred with water. The suspension was extracted with methylene chloride (3x) and the combined organic phases were dried over Na₂SO₄, concentrated and distilled. **10** (198 g, b.p.:95-100°C/0.2) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 3.67 (s, 3H); 3.62 (s, 2H); 2.28 (s, 6H).

Sodium hydride (60 g, 1.6 mol) was suspended in dimethyl carbonate (1500 ml, 17.7 mol) and heated to 90°C. **10** (198 g, 0.77 mol) dissolved in dimethyl carbonate (1200 ml, 14.2 M) was added dropwise to the reaction mixture. The reaction mixture was stirred at 90°C for 20 hours and cooled, and excess sodium hydride was destroyed with methanol. The reaction mixture was poured into ice/water, adjusted to pH = 5 with HCl, and extracted with methylene chloride (4x). The combined organic phases were washed with brine, dried over Na₂SO₄, concentrated and recrystallised from ethyl acetate/hexane (1:10). **11** (161.33 g, m.p.: 69-71°C) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 7.21 (s, 2H); 4.98 (s, 1H); 3.76 (s, 6H) 2.31 (s, 6H).

11 (40 g, 0.129 mol) and hydrazine **16** (27.15, 0.155 mol) were suspended in xylene (500 ml) and degassed for 0.3 hour by the introduction of argon into the reaction mixture. Triethylamine (43 ml, 0.1552 mol) was added to the reaction mixture, which was stirred for 4 hours at 140°C. The reaction mixture was cooled to room temperature and concentrated, and the residue was stirred with 10% HCl and extracted 3 times with ethyl acetate. The combined organic phases were washed with brine, dried over Na₂SO₄ and concentrated. **17** was obtained (42.87 g, m.p: 299-301°C). ¹H-NMR (300 MHz, CDCl₃): δ = 7.26 and 7.18 (2s, 2H); 4.70 (s, 1H); 4.20 – 3.75 (m, 8H); 2.41 (s, 3H) 2.08 (s, 3H).

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17 (42.87 g, 0.12 mol) was suspended in acetonitrile (400 ml), and triethylamine (29 ml, 0.206 mol) was added. The solution, which was now clear, was cooled to 10°C, and diethylcarbamoyl chloride (26.2 ml, 0.206 mol) was added. The reaction mixture was heated at reflux for 10 hours, cooled to room temperature, poured into ice/water and adjusted to pH = 5 with concentrated HCl. The reaction mixture was extracted several times with ethyl acetate. The combined organic phases were washed with brine, dried over Na₂SO₄ and concentrated. The resulting resin was stirred with ether. The crystals obtained **18** (47 g, m.p. 122°C) were filtered off. ¹H-NMR (300 MHz, CDCl₃): δ = 7.21 (s, 2H); 4.28 (m, 2H); 3.92 (m, 6H); 3.2 (m, 4H); 2.22 (s, 6H), 1.0 (m, 6H).

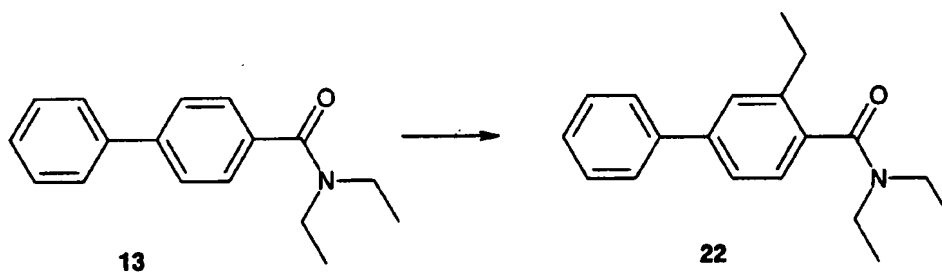
18 (2.76 g, 6.1 mmol), sodium carbonate (2.5 ml of a 2M solution in water, 9.98 mmol) and phenylboric acid (0.18 g, 8.88 mmol) were suspended in 1,2-diethoxyethane and degassed by the introduction of argon into the reaction mixture. After the addition of tetrakis-(triphenylphosphine)palladium (0.36 g, 0.312 mmol), the reaction mixture was stirred at reflux for 8 hours, cooled, poured into water and adjusted to pH = 5 with concentrated HCl. Extraction was carried out 3 times with ethyl acetate, and the combined organic phases were washed with brine, dried over Na₂SO₄, concentrated and chromatographed over silica gel (ethyl acetate:methanol = 5:1). **19** (2.7 g) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 7.7-7.25 (m, 7H); 4.3 (m, 2H); 3.92 (m, 6H); 3.2 (m, 4H); 2.29 (s, 6H), 0.95 (m, 6H).

19 (4.0 g, 8.9 mmol) was dissolved in methanol (50 ml), and sodium hydroxide solution (2N, 17.6 ml, 17.8 mmol) was added. The reaction mixture was stirred at reflux for 12 hours, cooled and concentrated. The residue was stirred with water and adjusted to a pH of 3 with concentrated HCl. The substance that precipitated was filtered off and dried. **20** (3.1 g, m.p.: 260-261°C) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 7.6-7.25 (m, 7H); 4.26 (m, 2H); 4.0 (m, 4H); 3.85 (m, 2H); 2.50 (s, 3H), 2.16 (s, 3H).

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20 (1.04 g, 2.96 mmol) was placed in acetonitrile (25 ml), and triethylamine (0.49 ml, 3.55 mmol) and pivalic acid chloride (0.45 ml, 3.55 mmol) were added. The reaction mixture was stirred at room temperature for 1 hour, concentrated, stirred with water and adjusted to a pH of 0.5 with concentrated HCl. Extraction was carried out three times with ethyl acetate, and the combined organic phases were washed with brine, dried over Na₂SO₄, concentrated and chromatographed over silica gel (ethyl acetate: methanol = 5:1). **21** (0.93 g, m.p. 159°C) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 7.6-7.25 (m, 7H); 4.3 (m, 2H); 3.94 (m, 2H); 3.85 (m, 4H); 2.28 (s, 6H), 1.05 (s, 9H).

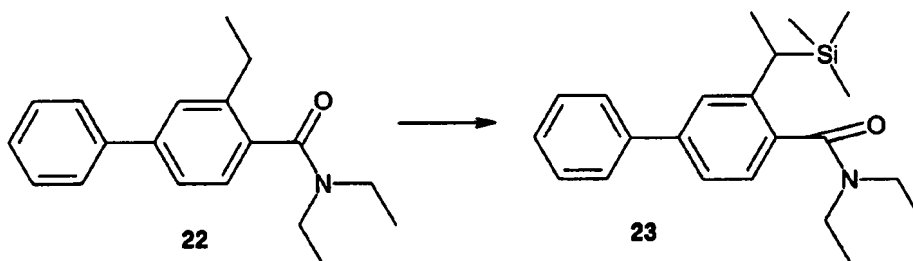
4-Biphenylcarbonyl chloride **12** (25 g, 0.115 mol) was dissolved in acetonitrile (150 ml), cooled to 0°C, and diethylamine (18.6 g, 0.254 mol) was added. The reaction mixture was stirred at room temperature for 0.5 hour, concentrated, taken up in ethyl acetate (200 ml) and washed with water (2 x 100 ml) and aqueous NaHCO₃ solution (100 ml), dried over Na₂SO₄ and concentrated. The crude product (28.5 g) was crystallised from Et₂O/hexane. **13** (24.5 g) was obtained. ¹H-NMR (300 MHz, CDCl₃): δ = 7.6-7.35 (m, 9H); 3.7-3.2 (2bs, 4H); 1.2 (m, 6H).



TMEDA (17.1 g, 0.147 mol) was dissolved in THF (distilled over Na, 500 ml), cooled to -78°C, and s-BuLi (113 ml of a 1.3M solution in cyclohexane, 0.147 mol) was added. The reaction mixture was stirred at -78°C for 0.25 hour, and a solution of **13** (33.9 g, 0.134 mol) in THF (100 ml) was added dropwise thereto, and the mixture was stirred at -78°C for a further 0.25 hour. The reaction mixture was treated dropwise with iodoethane (45.2 g, 0.29 mol) and heated to 0°C. An aqueous, saturated NH₄Cl solution (200 ml) was added to the reaction mixture. The organic phase was separated off, washed with brine, dried over Na₂SO₄, concentrated and chromatographed over silica gel (Et₂O/hexane = 1:1). **22** (29.5 g) was obtained.

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$^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.2 (m, 9H); 3.9-3.1 (m, 4H); 2.7 (q, 2H), 1.25 (t, 6H); 1.1 (t, 3H)



Diisopropylamine (13.36 g, 0.132 mol) was dissolved in THF (distilled over Na, 400 ml), cooled to -30°C and $n\text{-BuLi}$ (79 ml of a 1.6M solution in hexane, 0.126 mol) was added. After 0.25 hour, the reaction mixture was cooled to -78°C , and TMEDA (15.3 g, 0.132 mol) and **22** (29.5 g, 0.105 mol) dissolved in THF (75 ml) were slowly added. After 0.3 hour, chlorotrimethylsilylane was added to the reaction mixture, which was heated to 0°C . Aqueous, saturated NH_4Cl solution (200 ml) was added to the reaction mixture, and the organic phase was separated off. The organic phase was washed with brine, dried over Na_2SO_4 and concentrated. **23** (38.2) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.1 (m, 8H); 3.75 (m, 1H); 3.4-3.0 (m, 3H); 2.3 and 2.1 (2m, 1H); 1.35 (d, 3H); 1.25 (m, 6H); 0.0 (s, 9H).

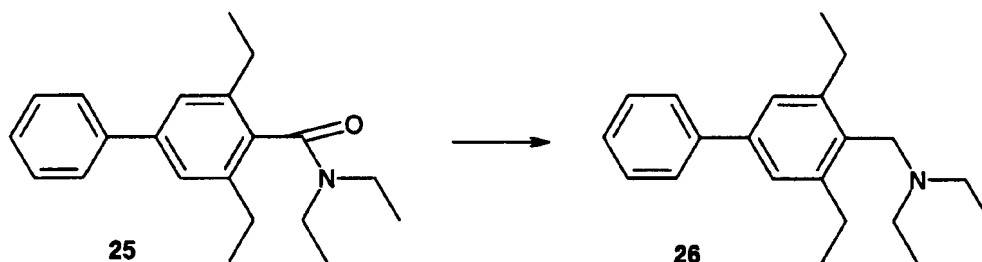
TMEDA (8.9 g, 0.077 mol) was dissolved in THF (distilled over Na, 400 ml), the reaction mixture was cooled to -78°C , and $s\text{-BuLi}$ (59 ml of a 1.3M solution in cyclohexane, 0.077 mol) and then a solution of **23** (24.8 g, 0.07 mol) in THF (75 ml) were added dropwise. After 0.5 hour at -78°C , iodomethane (13.3 g, 0.085 mol) was added to the reaction mixture, which was heated to 0°C . Aqueous, saturated NH_4Cl solution (200 ml) was added to the reaction mixture, and the organic phase was separated off. The organic phase was washed with brine, dried over Na_2SO_4 and concentrated. The crude product was chromatographed over silica gel (ethyl acetate/hexane = 1:7). **24** (18.5) was obtained. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.2 (m, 7H); 3.6 (m, 2H); 3.1 (m, 2H); 2.6 (m, 2H); 2.1 (m, 1H); 1.4 (d, 3H); 1.2 (m, 6H); 0.0 (s, 9H).

24 (18.5 g, 0.059 mol) was dissolved in DMF (100 ml) and water (10 ml), and CsF (11.2 g, 0.074 mol) was added at room temperature. The reaction mixture was stirred at 75°C for

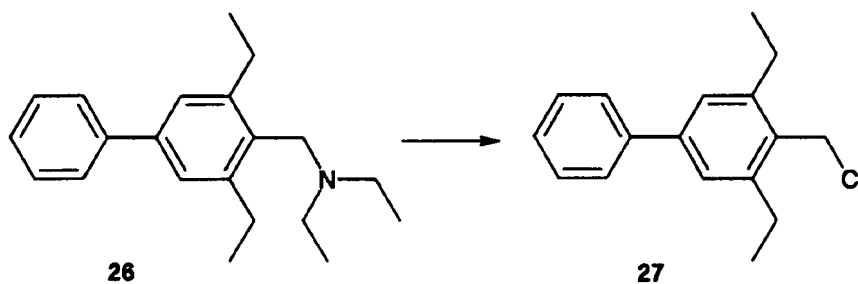
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8 hours, poured into water (300 ml) and extracted with ethyl acetate (2 x 100 ml). The combined organic phases were washed with brine, dried over Na_2SO_4 and concentrated.

25 (15.7 g) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.3 (m, 7H); 3.6 (m, 2H); 3.15 (m, 2H); 2.6 (m, 2H); 1.3 (m, 9H); 1.05 (t, 3H).

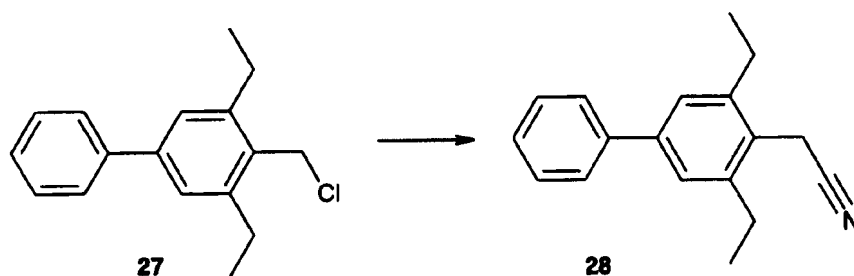


25 (15.7 g, 0.051 mol) was dissolved in toluene, and LiAlH_4 (1.92 g, 0.051 mol) was added. The reaction mixture was stirred at reflux for 20 hours and cooled to 0°C , HCl solution (1N, 150 ml) was added cautiously, and extraction was carried out with ethyl acetate (2 x 100 ml). The combined organic phases were extracted with 1N HCl solution (1 x 75 ml). The combined aqueous phases were adjusted to a pH of 11 with 2N NaOH solution, and extracted with ethyl acetate (2 x 100 ml). The combined organic phases were dried over Na_2SO_4 and concentrated. **26** (8.6 g) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.8-7.25 (m, 7H); 3.6 (s, 2H); 3.35 (q, 4H); 2.5 (q, 4H); 1.25 (t, 6H); 1.05 (t, 6H).

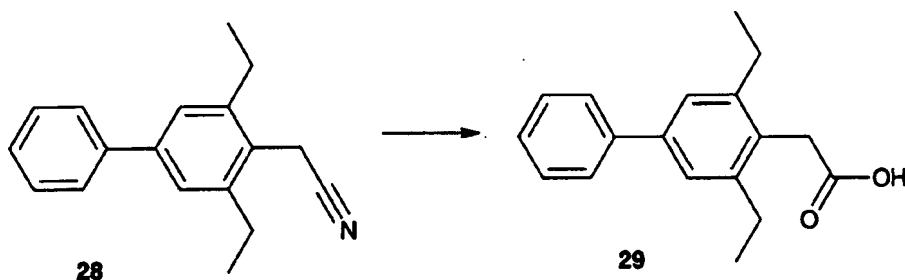


26 (8.6 g, 0.029 mol) was dissolved in toluene (50 ml), and chloroformic acid ethyl ester (12.6 g, 0.116 mol) was added. The reaction mixture was stirred at 75°C for 3 hours, and concentrated. **27** (11 g) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.3 (m, 7H); 4.75 (s, 2H); 2.9 (q, 4H); 1.3 (t, 6H).

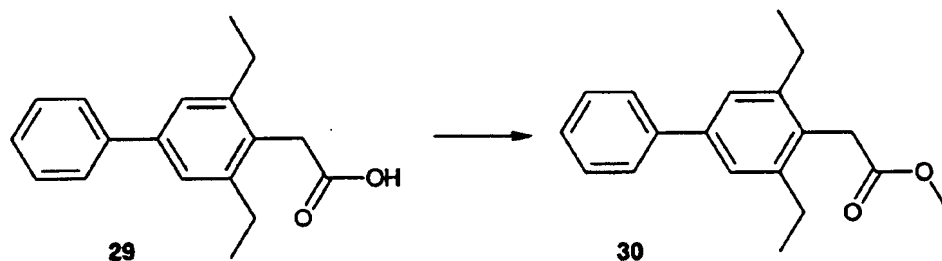
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27 (11 g of crude product, 0.029 mol) was stirred at reflux for 20 hours in acetonitrile (100 ml) together with finely triturated potassium cyanide (2.87 g, 0.044 mol). After the addition of additional potassium cyanide (2.87 g, 0.044 mol), the reaction mixture was stirred at reflux for a further 24 hours, cooled, poured into water (300 ml) and extracted with ethyl acetate (2 x 100 ml). The combined organic phases were washed with brine, dried over Na_2SO_4 , and concentrated. **28** (10 g) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300MHz, CDCl_3): δ = 7.6-7.3 (m, 7H); 3.7 (s, 2H); 2.8 (q, 4H); 1.4 (t, 6H).

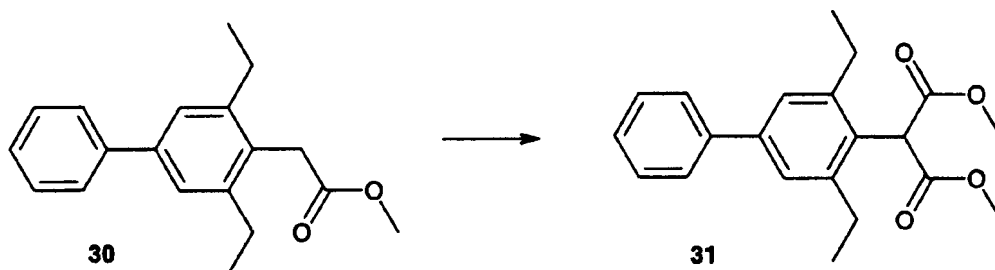


28 (4.5 g, 0.018 mol) was stirred in acetic acid (25 ml) and HCl (37% aqueous solution) for 24 hours at reflux. The acetic acid was distilled off, the reaction mixture was diluted with water (100 ml) and extracted with methylene chloride (2 x 100 ml). The combined organic phases were dried over Na_2SO_4 , and concentrated. **29** (5 g) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.3 (m, 7H); 3.8 (s, 2H); 2.7 (q, 4H); 1.25 (t, 6H).



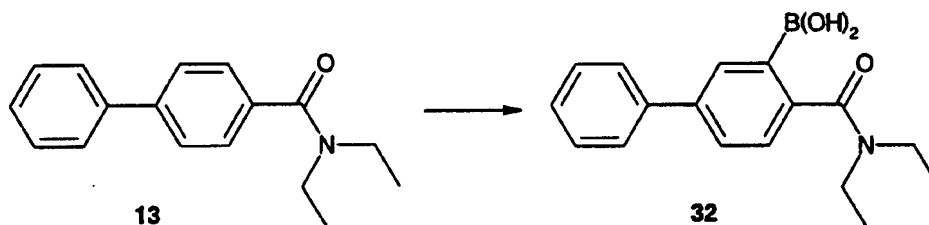
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29 (8.1 g, 0.03 mol) was dissolved in toluene (75 ml), and SOCl_2 (8.9 g, 0.075 mol) was added. The reaction mixture was heated to reflux temperature and, after the evolution of gas had ceased, concentrated. The resulting oil was taken up in toluene (50 ml), MeOH (20 ml) was added, and the mixture was stirred at room temperature for 0.5 hour and concentrated. **30** (9.1 g) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.3 (m, 7H), 3.8 (s, 2H); 3.7 (s, 3H); 2.7 (q, 4H); 1.25 (t, 6H).



30 (9.1 g, 0.032 mol) was dissolved in dimethyl carbonate (75 ml) and the solution was added to a suspension of sodium hydride (3.24 g of a 60% dispersion in mineral oil, 0.081 mol). The reaction mixture was stirred at reflux for 20 hours, cooled to room temperature, and 1N HCl solution (100 ml) was added slowly. After the evolution of gas had ceased, the reaction mixture was extracted with ethyl acetate (2 x 100 ml), and the combined organic phases were washed with brine, dried over Na_2SO_4 and concentrated.

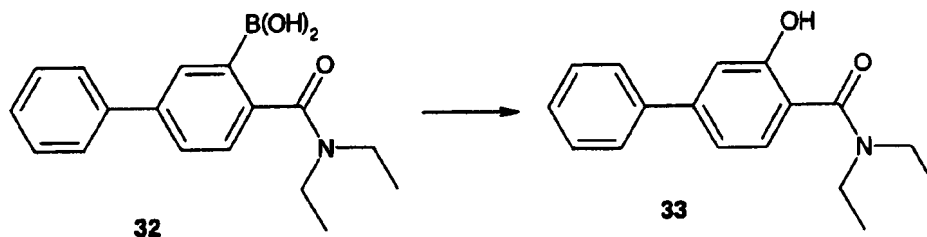
31 (11.3 g) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.2 (m, 7H); 5.1 (s, 1H); 3.7 (s, 6H); 2.7 (q, 4H); 1.2 (t, 6H).



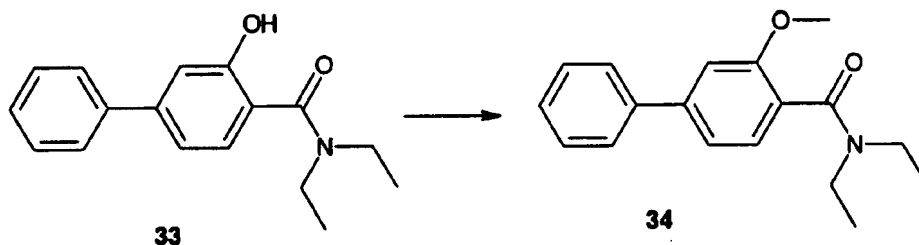
TMEDA (15.1 g, 0.13 mol) was dissolved in THF (500 ml) and cooled to -78°C , and $s\text{-BuLi}$ (100 ml of a 1.3M solution in cyclohexane, 0.13 mol) was added dropwise. **13** (30 g, 0.12 mol) was dissolved in THF (100 ml), and the solution was added dropwise to the reaction mixture over the course of 0.5 hour. After a further 0.5 hour at -78°C , trimethyl borate was added to the reaction mixture. The reaction mixture was stirred at -78°C for 0.5 hour, heated to -40°C , and an aqueous NH_4Cl solution (250 ml) was added thereto. The

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organic phase was washed with brine, dried over Na_2SO_4 and concentrated. **32** (36.7 g) was obtained in the form of a crude product.

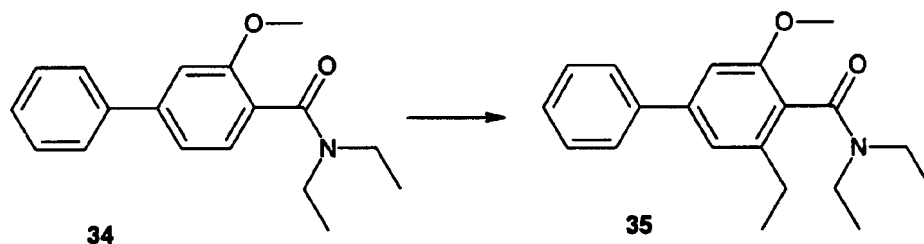


32 (35.2 g, 0.12 mol) was in MeOH (300 ml) and, with ice-cooling, H_2O_2 (16.1 g of a 30% solution, 0.142 mol) was added, the reaction temperature rising from 20°C to 40°C. After 2 hours, the reaction mixture was concentrated to 2/3, and seed crystals and water (500 ml) were added. The resulting crystals were filtered off and washed with water. **33** (30 g) was obtained. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.65-7.2 (m, 7H); 7.05 (dd, 1H); 3.05 (q, 4H); 1.3 (t, 6H).



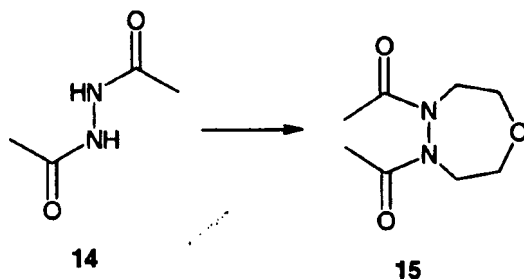
33 (3.7 g, 13.7 mmol) was placed in DMF, and NaH (0.55 g of a 60% dispersion, 13.7 mmol) was added. After the evolution of hydrogen had ceased, dimethyl sulfate (2.0 g, 16 mmol) was added to the reaction mixture, which was stirred at room temperature for 1 hour, poured into dilute HCl (300 ml) and extracted with ethyl acetate (2 x 100 ml). The combined organic phases were washed with brine, dried over Na_2SO_4 , concentrated and chromatographed over silica gel (ethyl acetate:hexane, 1:1). **34** (3.4 g) was obtained. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.2 (m, 8H); 3.9 (s, 3H); 3.6 (bm, 2H); 3.2 (q, 2H); 1.3 (t, 3H); 1.1 (t, 3H).

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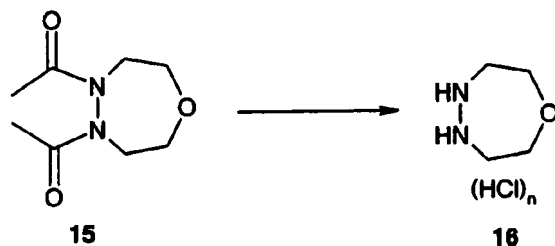
TMEDA (1.49 g, 12.8 mmol) was dissolved in THF (50 ml) and cooled to -78°C .

s-BuLi (9.8 ml of a 1.3M solution in cyclohexane, 12.8 mmol) was added to the reaction mixture, which was stirred at -78°C for 0.25 hour, a solution of **34** (3.3 g, 11.6 mmol) in THF (15 ml) was added dropwise and the mixture was stirred at -78°C for 0.3 hour. Ethyl iodide (2.0 g, 13 mmol) was added to the reaction mixture. The reaction mixture was heated to 0°C and an aqueous NH_4Cl solution (50 ml) was added. The organic phase was washed with brine, dried over Na_2SO_4 and concentrated. **35** (3.5 g) was obtained in the form of a crude product. $^1\text{H-NMR}$ (300 MHz, CDCl_3): δ = 7.6-7.3 (m, 5H); 7.07 (s, 1H); 6.9 (s, 1H); 3.9 (s, 3H); 3.8 (m, 1H); 3.45 (m, 1H) 3.2 (m, 2H); 2.6 (m, 2H); 1.3 (m, 6H); 1.1 (t, 3H).



N,N'-Diacetylhydrazine **14** (768 g, 6.62 mol), 2,2'-dichlorodiethyl ether (1141 g, 7.9 mol) and potassium carbonate (1827 g, 13.3 mol) were placed in DMF (8.5 litres) at room temperature. The resulting suspension was heated at 130°C for 3.5 hours, cooled, filtered and concentrated. The residue was taken up in toluene (1000 ml) and stirred at 0°C for 16 hours. After filtration, the product **15** was obtained in the form of white crystals (574 g). The mother liquor was concentrated by evaporation and recrystallised from a small amount of methanol. Again product **15** (108 g) was obtained in the form of white crystals.

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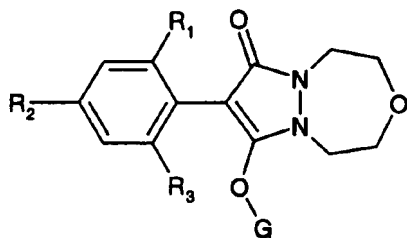
15 (1000 g, 5.38 mol) was dissolved in anhydrous methanol (5.4 litres) at 35°C, and acetyl chloride (1688 g, 21.5 mol) was added dropwise. After the addition was complete, the reaction mixture was stirred at 55°C for 16 hours. The reaction mixture was cooled and concentrated to about 1.4 litres, and stirred at from 0°C to 10°C. The crystals that formed in the process were filtered off and washed with ether (1.5 litres). The product **16** was obtained in the form of white crystals (520 g). ¹H-NMR (300 MHz, D₆-DMSO): δ = 3.8 (s, 4H); 3.4 (s, 4H).

The compounds listed in the following Tables were obtained in analogous manner.

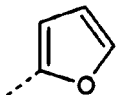
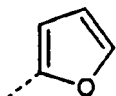


In the following Tables, "LC/MS: M⁺" denotes the molecular weight determined by coupled HPLC (High Performance Liquid Chromatography) and MS (Mass Spectrometry) analysis; the numbers given after "UV" indicate the frequency, in nanometres, of the absorption maxima of the UV spectrum measured in water/acetonitrile.


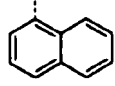
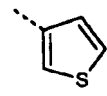
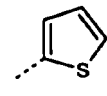
Table 1

Compounds of formula

**1a**

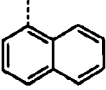




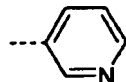
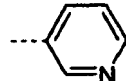
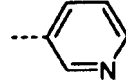
Comp. No.	R ₁	R ₂	R ₃	G	Phys. data
1a-1	CH ₃	Ph	CH ₃	H	m.p. 260-261°C
1a-2	CH ₃	Ph	CH ₃	C(O)C(CH ₃) ₃	m.p. 159-161°C
1a-3	CH ₃	Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	resin
1a-4	CH ₂ CH ₃	Ph	CH ₂ CH ₃	H	m.p. >250°C
1a-5	CH ₂ CH ₃	Ph	CH ₂ CH ₃	C(O)C(CH ₃) ₃	m.p. 162-164°C
1a-6	CH ₂ CH ₃	Ph	OCH ₃	H	m.p. 205-207°C
1a-7	CH ₂ CH ₃	Ph	OCH ₃	C(O)C(CH ₃) ₃	m.p. 57-59°C
1a-8	CH ₂ CH ₃	Ph	SCH ₃	H	m.p. 200-201°C
1a-9	CH ₂ CH ₃	Ph	SCH ₃	C(O)C(CH ₃) ₃	m.p. 197-199°C
1a-10	CH ₂ CH ₃	Ph	S(O) ₂ CH ₃	C(O)C(CH ₃) ₃	m.p. 172-174°C
1a-11	CH ₂ CH ₃	Ph	CH ₂ Ph	H	m.p. 203-205°C
1a-12	CH ₃	4-CH ₂ =CH-Ph	CH ₃	H	(LC/MS: M ⁺ = 376)
1a-13	CH ₃	2-Cl-Ph	CH ₃	H	(LC/MS: M ⁺ = 384)
1a-14	CH ₃	4-Cl-Ph	CH ₃	H	(LC/MS: M ⁺ = 384)
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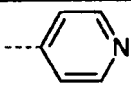
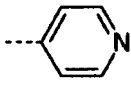
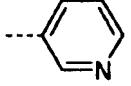
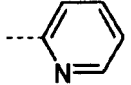
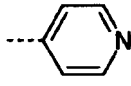
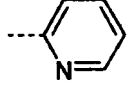
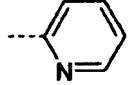
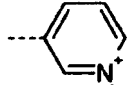
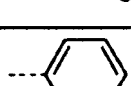
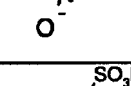
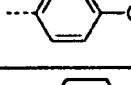

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Ia-18	CH ₃	4-CH ₃ O-Ph	CH ₃	H	(LC/MS: M ⁺ = 380)
Ia-19	CH ₃	3-CH ₃ CH ₂ O- Ph	CH ₃	H	(LC/MS: M ⁺ = 394)
Ia-20	CH ₃ CH ₂	3-CH ₃ CH ₂ O- Ph	CH ₃ CH ₂	H	(LC/MS: M ⁺ = 422)
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Ia-25	CH ₃ CH ₂		CH ₃ CH ₂	H	
Ia-26	CH ₃		CH ₃	H	(LC/MS: M ⁺ = 356)
Ia-27	CH ₂ CH ₃		CH ₂ CH ₃	H	(LC/MS: M ⁺ = 384)
Ia-28	CH ₂ CH ₃	2,4-Cl ₂ -Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 446)
Ia-29	CH ₂ CH ₃	4-F-Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 396)
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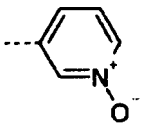
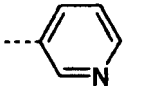
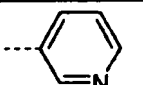
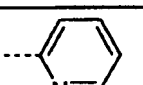
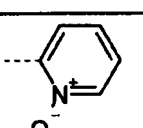
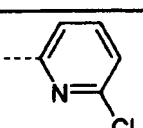
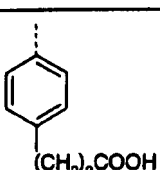
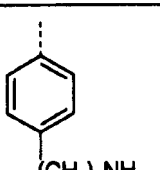
Ia-31	CH ₂ CH ₃	3,5-Cl ₂ -Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 446)
Ia-32	CH ₂ CH ₃	4-CF ₃ -Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 446)
Ia-33	CH ₂ CH ₃		CH ₃	C(O)C(CH ₃) ₃	m.p. 139-141°C
Ia-34	CH ₃	4-CH ₂ =CH-Ph	CH ₃	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 475)
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Ia-36	CH ₃		CH ₃	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 499)
Ia-37	CH ₃	4-Cl-Ph	CH ₃	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 483)
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Ia-44	CH ₃		CH ₃	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 455)
Ia-45	CH ₃		CH ₃	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 455)

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Ia-46	CH ₃		CH ₃	H	(LC/MS: M ⁺ = 400)
Ia-47	CH ₃		CH ₃	H	(LC/MS: M ⁺ = 356)
Ia-48	CH ₃ CH ₂	4-Cl-Ph	CH ₃ CH ₂	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 511)
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Ia-51	CH ₃ CH ₂		CH ₃ CH ₂	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 483)
Ia-52	CH ₃ CH ₂		CH ₃ CH ₂	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 483)
Ia-53	CH ₂ CH ₃	3-Cl,4-F-Ph	CH ₂ CH ₃	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 529)
Ia-54	CH ₃ CH ₂	3,5-Cl ₂ -Ph	CH ₃ CH ₂	C(O)N- (CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 545)
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Ia-56	CH ₂ CH ₃		CH ₂ CH ₃	H	(LC/MS: M ⁺ = 384)
Ia-57	CH ₃		CH ₃	C(O)N- (CH ₂ CH ₃) ₂	solid
Ia-58	CH ₃		CH ₃	H	solid
Ia-59	CH ₃		CH ₃	C(O)C(CH ₃) ₃	solid

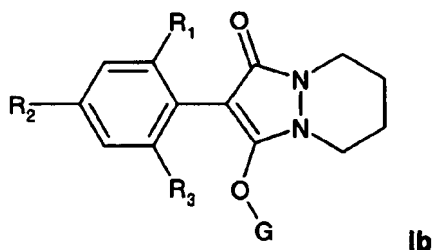
la-60	CH ₃		CH ₃	H	solid
la-61	CH ₃		CH ₃	C(O)C(CH ₃) ₃	m.p. 99-100°C
la-62	CH ₂ CH ₃		CH ₂ CH ₃	C(O)C(CH ₃) ₃	wax
la-63	CH ₃		CH ₃	H	solid
la-64	CH ₂ CH ₃		CH ₂ CH ₃	C(O)C(CH ₃) ₃	amorphous
la-65	CH ₂ CH ₃		CH ₂ CH ₃	C(O)C(CH ₃) ₃	m.p. 146-147°C
la-66	CH ₂ CH ₃		CH ₂ CH ₃	H	m.p. 250°C
la-67	CH ₃		CH ₃	H	m.p. 250°C
la-68	CH ₂ CH ₃		CH ₂ CH ₃	C(O)C(CH ₃) ₃	
la-69	CH ₂ CH ₃		CH ₂ CH ₃	H	crystalline
la-70	CH ₃		CH ₃	C(O)N-(CH ₂ CH ₃) ₂	
la-71	CH ₃		CH ₃	C(O)N-(CH ₂ CH ₃) ₂	

la-72	CH ₃		CH ₃	C(O)N-(CH ₂ CH ₃) ₂	
la-73	CH ₂ CH ₃		CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	
la-74	CH ₂ CH ₃		CH ₂ CH ₃	H	
la-75	CH ₃		CH ₃	C(O)C(CH ₃) ₃	
la-76	CH ₂ CH ₃		CH ₂ CH ₃	H	m.p. 250°C
la-77	CH ₂ CH ₃		CH ₂ CH ₃	C(O)C(CH ₃) ₃	
la-78	CH ₃		CH ₃	H	m.p. 284-286°C
la-79	CH ₃		CH ₃	H	m.p. 230°C (decomp.)

Ph = phenyl

Table 2





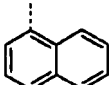
Compounds of formula

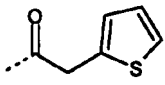
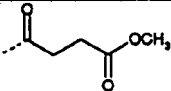
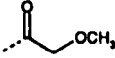
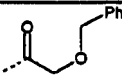
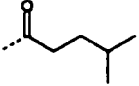
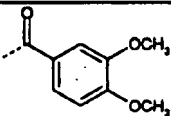
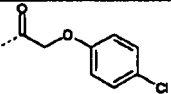

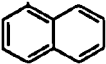






Comp. No.	R ₁	R ₂	R ₃	G	Phys. data
Ib-1	CH ₃	Ph	CH ₃	H	m.p. 239-240°C
Ib-2	CH ₃	Ph	CH ₃	C(O)C(CH ₃) ₃	resin
Ib-3	CH ₃	Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	resin
Ib-4	CH ₃	Ph	CH ₃	C(O)O-CH ₂ CH ₃	m.p. 144-146°C
Ib-5	CH ₃	Ph	CH ₃	C(O)CH ₃	m.p. 198-199°C
Ib-6	CH ₃	Ph	CH ₃	C(O)cyclopropyl	m.p. 104-105
Ib-7	H	Ph	CH ₂ CH ₃	H	m.p. 176-177°C
Ib-8	H	Ph	CH ₂ CH ₃	C(O)C(CH ₃) ₃	m.p. 128-130°C
Ib-9	CH ₂ CH ₃	Ph	CH ₂ CH ₃	H	m.p. >250°C
Ib-10	CH ₂ CH ₃	Ph	CH ₂ CH ₃	C(O)C(CH ₃) ₃	m.p. 166-168°C
Ib-11	CH ₂ CH ₃	Ph	SCH ₃	H	m.p. 170-172°C
Ib-12	CH ₂ CH ₃	Ph	SCH ₃	C(O)C(CH ₃) ₃	m.p. 182-183°C
Ib-13	CH ₂ CH ₃	Ph	OCH ₃	H	m.p. 148-150°C
Ib-14	CH ₂ CH ₃	Ph	OCH ₃	C(O)C(CH ₃) ₃	m.p. 127-129°C
Ib-15	CH ₂ CH ₃	Ph	C≡CSi(CH ₃) ₃	H	m.p. 150-152°C
Ib-16	CH ₂ CH ₃	Ph	C≡CSi(CH ₃) ₃	C(O)C(CH ₃) ₃	m.p. 69-71°C
Ib-17	CH ₂ CH ₃	Ph	C≡CSi(CH ₃) ₃	C(O)N-(CH ₂ CH ₃) ₂	m.p. 134-136°C
Ib-18	CH ₂ CH ₃	Ph	C≡CH	H	m.p. 108-111°C
Ib-19	CH ₂ CH ₃	Ph	C≡CH	C(O)C(CH ₃) ₃	m.p. 134-136°C
Ib-20	CH ₂ CH ₃	Ph	Br	H	m.p. 221-222°C
Ib-21	CH ₂ CH ₃	Ph	Br	C(O)C(CH ₃) ₃	m.p. 153-154°C




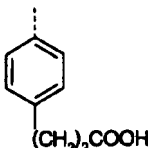
lb-22	CH ₂ CH ₃	Ph	Br	C(O)N-(CH ₂ CH ₃) ₂	m.p. 129-131°C
lb-23	CH ₂ CH ₃	Ph	CH ₂ Ph	H	m.p. 247-249°C
lb-24	CH ₂ CH ₃	Ph	CH ₂ Ph	C(O)C(CH ₃) ₃	resin
lb-25	CH ₃	3-CF ₃ -Ph	CH ₃	H	m.p. 225-226°C
lb-26	CH ₃	3-CF ₃ -Ph	CH ₃	C(O)C(CH ₃) ₃	m.p. 197-199°C
lb-27	CH ₃	3-CF ₃ -Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	oil
lb-28	CH ₃	3-CF ₃ -Ph	CH ₃	C(O)O-CH ₂ CH ₃	resin
lb-29	CH ₃	4-F-Ph	CH ₃	H	
lb-30	CH ₃	4-F-Ph	CH ₃	C(O)C(CH ₃) ₃	
lb-31	CH ₃	4-F-Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	oil
lb-32	CH ₃	4-CF ₃ -Ph	CH ₃	H	m.p. 294-296°C
lb-33	CH ₃	4-CF ₃ -Ph	CH ₃	C(O)C(CH ₃) ₃	m.p. 160-161°C
lb-34	CH ₃	4-CF ₃ -Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	m.p. 68-70°C
lb-35	CH ₃	4-CF ₃ -Ph	CH ₃	C(O)OCH ₂ CH ₃	resin
lb-36	CH ₃	3-NO ₂ -Ph	CH ₃	H	m.p. 219-221°C
lb-37	CH ₃	3-NO ₂ -Ph	CH ₃	C(O)C(CH ₃) ₃	m.p. 180-182°C
lb-38	CH ₃	3-NO ₂ -Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	
lb-39	CH ₃	4-CH ₂ =CH-Ph	CH ₃	H	(LC/MS: M* = 360)
lb-40	CH ₃	2-Cl-Ph	CH ₃	H	(LC/MS: M* = 368)
ld-41	CH ₃	4-Cl-Ph	CH ₃	H	(LC/MS: M* = 368)
lb-42	CH ₂ CH ₃	4-Cl-Ph	CH ₂ CH ₃	H	(LC/MS: M* = 396)
lb-43	CH ₃	3-Br-Ph	CH ₃	H	(LC/MS: M* = 412)
lb-44	CH ₃	2-CH ₃ O-Ph	CH ₃	H	(LC/MS: M* = 364)

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Ib-45	CH ₃	4-CH ₃ O-Ph	CH ₃	H	(LC/MS: M ⁺ = 364)
Ib-46	CH ₃	3-CH ₃ CH ₂ O-Ph	CH ₃	H	(LC/MS: M ⁺ = 378)
Ib-47	CH ₃ CH ₂	3-CH ₃ CH ₂ O-Ph	CH ₃ CH ₂	H	(LC/MS: M ⁺ = 406)
Ib-48	CH ₃	4-CH ₃ S-Ph	CH ₃	H	(LC/MS: M ⁺ = 380)
Ib-49	CH ₃	2-CH ₃ -Ph	CH ₃	H	(LC/MS: M ⁺ = 348)
Ib-50	CH ₃	4-CH ₃ -Ph	CH ₃	H	(LC/MS: M ⁺ = 348)
Ib-51	CH ₃		CH ₃	H	
Ib-52	CH ₃ CH ₂		CH ₃ CH ₂	H	
Ib-53	CH ₃		CH ₃	H	(LC/MS: M ⁺ = 340)
Ib-54	CH ₃ CH ₂		CH ₃	H	
Ib-55	CH ₃		CH ₃	H	(LC/MS: M ⁺ = 384)
Ib-56	CH ₂ CH ₃	2,4-Cl ₂ -Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 430)
Ib-57	CH ₂ CH ₃	4-F-Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 380)
Ib-58	CH ₂ CH ₃	3-Cl,4-F-Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 414)
Ib-59	CH ₂ CH ₃	3,5-Cl ₂ -Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 430)

Ib-60	CH ₂ CH ₃	4-CF ₃ -Ph	CH ₂ CH ₃	H	(LC/MS: M ⁺ = 430)
Ib-61	CH ₃	Ph	CH ₃		UV: 212.1 nm 270.9 nm
Ib-62	CH ₃	Ph	CH ₃		UV: 209.7 nm 270.9 nm
Ib-63	CH ₃	Ph	CH ₃		UV: 207.4 nm 270.9 nm
Ib-64	CH ₃	Ph	CH ₃		UV: 207.4 nm 270.9 nm
Ib-65	CH ₃	Ph	CH ₃	C(O) <i>n</i> C ₇ H ₁₅	UV: 205.0 nm 270.9 nm
Ib-66	CH ₃	Ph	CH ₃		UV: 207.4 nm 268.5 nm
Ib-67	CH ₃	Ph	CH ₃		UV: 205.0 nm 270.9 nm
Ib-68	CH ₃	Ph	CH ₃		UV: 270.9 nm
Ib-69	CH ₃	Ph	CH ₃		
Ib-70	CH ₃	2-Cl-Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 467)
Ib-71	CH ₃		CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 483)

lb-72	CH ₃	4-Cl-Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 467)
lb-73	CH ₃	2-CH ₃ O-Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 463)
lb-74	CH ₃	4-CH ₃ S-Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 479)
lb-75	CH ₃	4-CH ₃ O-Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 463)
lb-76	CH ₃	3-Br-Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 511)
lb-77	CH ₃	3-CH ₂ CH ₃ O-Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 477)
lb-78	CH ₃	2-CH ₃ -Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 447)
lb-79	CH ₃	4-CH ₃ -Ph	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 447)
lb-80	CH ₃		CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 439)
lb-81	CH ₃		CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 439)
lb-82	CH ₃		CH ₃	H	(LC/MS: M ⁺ = 340)
lb-83	CH ₂ CH ₃	4-Cl-Ph	CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 495)
lb-84	CH ₂ CH ₃	3-CH ₃ CH ₂ O-Ph	CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 505)
lb-85	CH ₂ CH ₃	4-F-Ph	CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 479)
lb-86	CH ₂ CH ₃		CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 467)

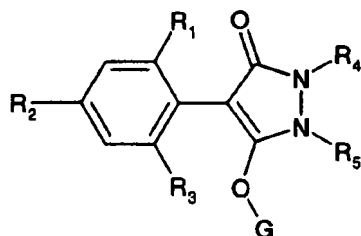
lb-87	CH ₂ CH ₃		CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 467)
lb-88	CH ₂ CH ₃	3-Cl,4-F-Ph	CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 513)
lb-89	CH ₂ CH ₃	3,5-Cl ₂ -Ph	CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 529)
lb-90	CH ₂ CH ₃	2,4-Cl ₂ -Ph	CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	(LC/MS: M ⁺ = 529)
lb-91	CH ₂ CH ₃	4-CF ₃ -Ph	CH ₂ CH ₃	C(O)N-(CH ₂ CH ₃) ₂	
lb-92	CH ₂ CH ₃		CH ₂ CH ₃	H	(LC/MS: M ⁺ = 368)
lb-93	CH ₂ CH ₃		CH ₂ CH ₃	H	(LC/MS: M ⁺ = 368)
lb-94	CH ₃		CH ₃	H	m.p. 224-226°C

Ph = phenyl

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Table 3

Compounds of formula

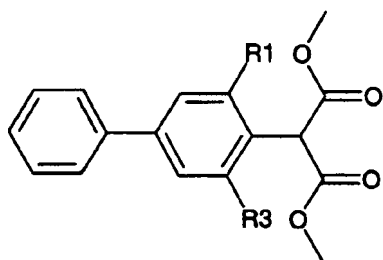
**Ic**

Comp. No.	R ₁	R ₂	R ₃	R ₄	R ₅	G	Phys. data
Ic-1	CH ₃	Ph	CH ₃	CH ₃	CH ₃	C(O)N-(CH ₂ CH ₃) ₂	resin
Ic-2	CH ₂ CH ₃	Ph	OCH ₃	H	(CH ₂) ₃ OH	H	m.p. 91-93°C
Ic-3	CH ₃	Ph	CH ₃	CH ₃	CH ₃	H	m.p. 197°C

Ph = phenyl

Table 4

Compounds of formula

**Id**

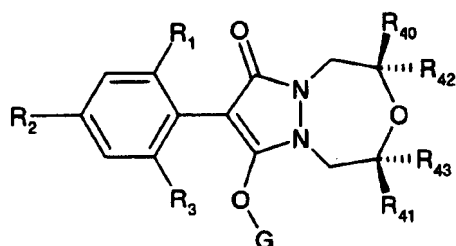
Comp. No.	R ₁	R ₃	Phys. data
Id-1	CH ₃	CH ₃	m.p. 68-69°C

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Id-2	CH ₂ CH ₃	SCH ₃	oil
Id-3	CH ₂ CH ₃	OCH ₃	oil
Id-4	CH ₂ CH ₃	CH ₂ CH ₃	oil
Id-5	CH ₂ CH ₃	CH ₂ Ph	oil
Id-6	CH ₂ CH ₃	Br	oil
Id-7	CH ₂ CH ₃	C≡CSi(CH ₃) ₃	oil
Id-8	CH ₂ CH ₃	H	oil

Ph = phenyl

Table 5



1e

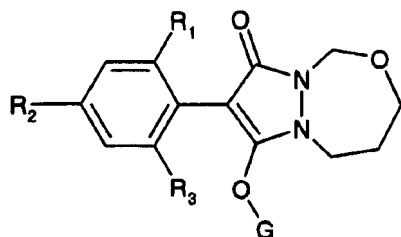
Comp. No.	R ₁	R ₂	R ₃	G	R ₄₀	R ₄₁	R ₄₂	R ₄₃	Phys. data
1e-1	CH ₃ CH ₂	Ph	CH ₃ O	H	H	H	-(CH ₂) ₂ -		m.p. 196-197°C
1e-2	CH ₃ CH ₂	Ph	CH ₃ O	H	CH ₃ CH ₂	H	H	H	m.p. 133-135°C
1e-3	CH ₃ CH ₂	Ph	CH ₃ O	H	CH ₃	CH ₃	H	H	m.p. 139-145°C
1e-4	CH ₃ CH ₂	Ph	CH ₃ O	H	CH ₃	CH ₃	H	H	m.p. 119°C
1e-5	CH ₃ CH ₂	Ph	CH ₃ O	C(O)- C(CH ₃) ₃	CH ₃ CH ₂	H	H	H	crystalline

Ph = phenyl

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Table 6

Compounds of formula

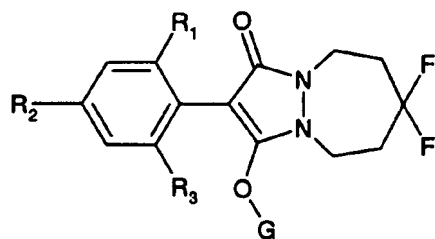
**If**

Comp. No.	R ₁	R ₂	R ₃	G	Phys. data
If-1	CH ₃ CH ₂	Ph	CH ₃ O	H	m.p. 184-188°C

Ph = phenyl

Table 7

Compounds of formula

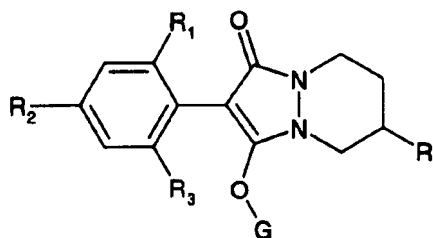
**Ig**

Comp. No.	R ₁	R ₂	R ₃	G	Phys. data
Ig-1	CH ₃ CH ₂	Ph	CH ₃ O	H	m.p. 147-149°C

Ph = phenyl

Table 8

Compounds of formula



1h

Comp No.	R ₁	R ₂	R ₃	R	G	Phys. data
1h-1	-OCH ₃	Ph	C ₂ H ₅	OH	C(O)C(CH ₃) ₃	¹ H NMR (CDCl ₃ , 300 MHz) δ = 7.6 (d, 2H); 7.42 (t, 2H); 7.34 (t, 1H); 7.1 (s, 1H); 6.88 (s, 1H); 4.2 - 3.14 (m, 6H); 3.8 (2s, 3H); 2.67 (m, 2H); 2.05 (m, 2H); 1.2 (m, 3H); 1.1 (m, 9H) (Mixture of isomers)
1h-2	CH ₂ CH ₃	Ph	OCH ₃	H ₃ C-O-CH ₂ CH ₂ -O-	C(O)C(CH ₃) ₃	¹ H NMR (CDCl ₃ , 300 MHz) δ = 7.6 (d, 2H); 7.42 (t, 2H); 7.34 (t, 1H); 7.1 (s, 1H); 6.88 (s, 1H); 4.5 (m, 0.26H); 4.2 (m,

PH/5-31143A

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						0.17H); 4.0 - 3.3 (m, 8H); 3.8 (bs, 3H);, 3.4 (2s, 3H); 3.2 (m, 0.5H); 2.7 (m, 2H), 2.3 - 1.85 (m, 2H), 1.2 (m, 3H), 1.1 (m, 9H) (Mixture of isomers)
lh-3	CH ₂ CH ₃	Ph	CH ₂ -CH ₃	H ₃ C-O-CH ₂ CH ₂ -O-	C(O)C(CH ₃) ₃	
lh-4	CH ₂ CH ₃	Ph	ethynyl	H ₃ C-O-CH ₂ CH ₂ -O-	C(O)C(CH ₃) ₃	
lh-5	CH ₂ CH ₃	Ph	CH ₂ -CH ₃	H ₃ C-CH ₂ -O-CH ₂ CH ₂ -O-	C(O)C(CH ₃) ₃	
lh-6	CH ₃	Ph	CH ₃	H ₃ C-O-CH ₂ CH ₂ -O-	C(O)C(CH ₃) ₃	

Ph = phenyl

Example B2: Postemergence herbicidal action

Monocotyledonous and dicotyledonous test plants are sown in standard soil in pots. At the 2- to 3-leaf stage of the test plants, an aqueous suspension (prepared from a wettable powder (Example F3, b) according to WO 97/34485) of the test compound or an emulsion (prepared from an emulsifiable concentrate (Example F1, c) according to WO 97/34485) of the test compound is applied by spraying at an optimum concentration (500 litres of water/ha). The test plants are then cultivated further in a greenhouse under optimum conditions.

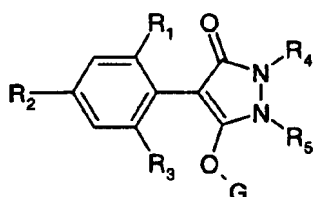
After a test duration of from 2 to 3 weeks, the test is evaluated in accordance with a scale of nine ratings (1 = total damage, 9 = no action). Ratings of from 1 to 4 (especially from 1 to 3) indicate good to very good herbicidal action.

Table B2:

Compound	Rate of application (g/ha)	Avena	Lollum	Setaria	Sinapis
Ib-1	2000	1	2	1	2
Ib-4	2000	1	3	1	2
Ib-2	2000	3	3	1	2
Ia-33	2000	2	2	1	2
Ib-66	2000	1	2	1	2
Ia-4	2000	1	1	1	2
Ia-5	2000	1	1	1	2
Ib-9	2000	1	1	1	2
Ib-10	2000	1	1	1	2
Ia-1	2000	1	1	1	2
Ia-2	2000	1	1	1	2
Ia-6	2000	1	1	1	2
Ia-7	2000	1	1	1	2
Ib-13	2000	1	1	1	2
Ib-14	2000	1	1	1	2
Ib-28	2000	1	2	1	2
Ia-8	2000	2	1	1	2
Ib-20	2000	2	2	1	2
Ib-21	2000	1	1	1	2
Ib-69	2000	2	2	1	2
Ib-18	2000	1	1	1	2
Ib-19	2000	1	1	1	2
Ie-1	2000	1	1	1	2
If-1	2000	1	1	1	3

In that test, the compounds of formula I exhibit strong herbicidal action.

The same results are obtained when the compounds of formula I are formulated according to the other Examples of WO 97/34485.

Patent claims:**1. A compound of formula I**

I,

wherein

R₁ and R₃ are each independently of the other hydrogen, halogen, nitro, cyano, C₁-C₄alkyl, C₂-C₄alkenyl, C₂-C₄alkynyl, tri(C₁-C₄alkylsilyl)-C₂-C₄alkynyl, C₁-C₄haloalkyl, C₂-C₆haloalkenyl, C₃-C₆cycloalkyl, halo-substituted C₃-C₆cycloalkyl, benzyl, C₂-C₆alkoxyalkyl, C₂-C₆alkylthioalkyl, hydroxy, mercapto, C₁-C₆alkoxy, C₃-C₆alkenyloxy, C₃-C₆alkynyloxy, C₁-C₄alkylcarbonyl, C₁-C₄alkoxycarbonyl, C₁-C₄alkylthio, C₁-C₄alkylsulfinyl, C₁-C₄alkylsulfonyl, amino, C₁-C₄alkylamino, di(C₁-C₄alkyl)amino, C₁-C₄hydroxyalkyl, formyl, C₁-C₄alkylcarbonylamino or C₁-C₄alkylsulfonylamino,

R₂ is phenyl, naphthyl or a 5- or 6-membered aromatic ring that may contain 1 or 2 hetero atoms selected from the group nitrogen, oxygen and sulfur, it being possible for the phenyl ring, the naphthyl ring and the 5- or 6-membered aromatic ring to be substituted by halogen, C₃-C₆cycloalkyl, hydroxy, mercapto, amino, amino-C₁-C₆alkyl, carboxyl-C₁-C₆alkyl, cyano, nitro or by formyl; and/or

for the phenyl ring, the naphthyl ring and the 5- or 6-membered aromatic ring to be substituted by C₁-C₆alkyl, C₁-C₆alkoxy, hydroxy-C₁-C₆alkyl, C₁-C₆alkoxy-C₁-C₆alkyl, C₁-C₆alkoxy-C₁-C₆alkoxy, C₁-C₆alkylcarbonyl, C₁-C₆alkylthio, C₁-C₆alkylsulfinyl, C₁-C₆alkylsulfonyl, mono-C₁-C₆alkylamino, di-C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, C₁-C₆alkylcarbonyl-(C₁-C₆alkyl)amino, C₂-C₆alkenyl, C₃-C₆alkenyloxy, hydroxy-C₃-C₆alkenyl, C₁-C₆alkoxy-C₃-C₆alkenyl, C₁-C₆alkoxy-C₃-C₆alkenyloxy, C₂-C₆alkenylcarbonyl, C₂-C₆alkenylthio, C₂-C₆alkenylsulfinyl, C₂-C₆alkenylsulfonyl, mono- or di-C₂-C₆alkenylamino, C₁-C₆alkyl-(C₃-C₆alkenyl)-amino, C₂-C₆alkenylcarbonylamino, C₂-C₆alkenylcarbonyl-(C₁-C₆alkyl)amino, C₂-C₆alkynyl, C₃-C₆alkynyloxy, hydroxy-C₃-C₆alkynyl, C₁-C₆alkoxy-C₃-C₆alkynyl, C₁-C₆alkoxy-C₄-C₆alkynyloxy, C₂-C₆alkynylcarbonyl, C₂-C₆alkynylthio, C₂-C₆alkynylsulfinyl, C₂-C₆alkynylsulfonyl,

mono- or di-C₃-C₆alkynylamino, C₁-C₆alkyl-(C₃-C₆alkynyl)amino, C₂-C₆alkynylcarbonylamino or by C₂-C₆alkynylcarbonyl-(C₁-C₆alkyl)amino; and/or

for the phenyl ring, the naphthyl ring and the 5- or 6-membered aromatic ring to be substituted by halo-substituted C₁-C₆alkyl, halo-substituted C₁-C₆alkoxy, halo-substituted hydroxy-C₁-C₆alkyl, halo-substituted C₁-C₆alkoxy-C₁-C₆alkyl, halo-substituted C₁-C₆alkoxy-C₁-C₆alkoxy, halo-substituted C₁-C₆alkylcarbonyl, halo-substituted C₁-C₆alkylthio, halo-substituted C₁-C₆alkylsulfinyl, halo-substituted C₁-C₆alkylsulfonyl, halo-substituted mono-C₁-C₆alkylamino, halo-substituted di-C₁-C₆alkylamino, halo-substituted C₁-C₆alkylcarbonylamino, halo-substituted C₁-C₆alkylcarbonyl-(C₁-C₆alkyl)amino, halo-substituted C₂-C₆alkenyl, halo-substituted C₃-C₆alkenyloxy, halo-substituted hydroxy-C₃-C₆alkenyl, halo-substituted C₁-C₆alkoxy-C₂-C₆alkenyl, halo-substituted C₁-C₆alkoxy-C₃-C₆alkenyloxy, halo-substituted C₂-C₆alkenylcarbonyl, halo-substituted C₂-C₆alkenylthio, halo-substituted C₂-C₆alkenylsulfinyl, halo-substituted C₂-C₆alkenylsulfonyl, halo-substituted mono- or di-C₃-C₆alkenylamino, halo-substituted C₁-C₆alkyl-(C₃-C₆alkenyl)amino, halo-substituted C₂-C₆alkenylcarbonylamino, halo-substituted C₂-C₆alkenylcarbonyl-(C₁-C₆alkyl)amino, halo-substituted C₂-C₆alkynyl, halo-substituted C₃-C₆alkynyloxy, halo-substituted hydroxy-C₃-C₆alkynyl, halo-substituted C₁-C₆alkoxy-C₃-C₆alkynyl, halo-substituted C₁-C₆alkoxy-C₄-C₆alkynyloxy, halo-substituted C₂-C₆alkynylcarbonyl, halo-substituted C₂-C₆alkynylthio, halo-substituted C₂-C₆alkynylsulfinyl, halo-substituted C₂-C₆alkynylsulfonyl, halo-substituted mono- or di-C₃-C₆alkynylamino, halo-substituted C₁-C₆alkyl-(C₃-C₆alkynyl)amino, halo-substituted C₂-C₆alkynylcarbonylamino or by halo-substituted C₂-C₆alkynylcarbonyl(C₁-C₆alkyl)amino; and/or

for the phenyl ring, the naphthyl ring and the 5- or 6-membered aromatic ring to be substituted by a radical of formula COOR₅₀, CONR₅₁, SO₂NR₅₃R₅₄ or SO₂OR₅₅, wherein R₅₀, R₅₁, R₅₂, R₅₃, R₅₄ and R₅₅ are each independently of the others hydrogen, C₁-C₆alkyl, C₂-C₆alkenyl or C₃-C₆alkynyl, or C₁-C₆alkyl, C₂-C₆alkenyl or C₃-C₆alkynyl each substituted by halogen, hydroxy, alkoxy, mercapto, amino, cyano, nitro, alkylthio, alkylsulfinyl or by alkylsulfonyl,

R₄ and R₅ are each independently of the other hydrogen, C₁-C₁₂alkyl, C₁-C₁₂haloalkyl, C₁-C₁₂hydroxyalkyl, C₃-C₆alkenyl, C₃-C₆alkynyl, C₁-C₁₀alkoxy-C₁-C₆alkyl, or C₃-C₆alkyl that may contain one or two oxygen atoms, C₁-C₁₀alkylthio-C₁-C₆alkyl, C₃-C₆cycloalkyl, C₃-C₆cycloalkyl that contains 1 or 2 hetero atoms selected from the group oxygen and sulfur,

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C₃-C₈halocycloalkyl, C₃-C₈halocycloalkyl that contains 1 or 2 hetero atoms selected from the group oxygen and sulfur, phenyl, or phenyl substituted by halogen, C₁-C₆alkyl, C₁-C₆haloalkyl, C₁-C₆alkoxy, C₁-C₆haloalkoxy, nitro or by cyano, or R₄ and R₅ are each independently of the other a 5- or 6-membered ring that may contain hetero atoms selected from the group oxygen, sulfur and nitrogen, or

R₄ and R₅, together with the atoms to which they are bonded, form a 5- to 8-membered ring, which may contain 1 or 2 oxygen atoms, sulfur atoms or NR₆ groups, wherein

R₆ is hydrogen, C₁-C₄alkyl, C₁-C₆alkylcarbonyl, C₁-C₆alkylsulfonyl, C₃-C₆alkenyl or C₃-C₆alkynyl, and which may be substituted by halogen, hydroxy, C₁-C₁₀alkyl, C₁-C₁₀alkoxy, C₁-C₁₀haloalkyl, C₃-C₈cycloalkyl, phenyl or by benzyl; or

which may be substituted by phenyl substituted by halogen, C₁-C₆alkyl, C₁-C₆haloalkyl, C₃-C₈cycloalkyl, hydroxy, C₁-C₆alkoxy, C₁-C₆alkoxy-C₁-C₆alkoxy, C₁-C₆haloalkoxy or by nitro, or by benzyl substituted by halogen, C₁-C₆alkyl, C₁-C₆haloalkyl, C₃-C₈cycloalkyl, hydroxy, C₁-C₆alkoxy, C₁-C₆haloalkoxy or by nitro; or

which may be substituted by CH₂-heteroaryl, wherein the aryl moiety has 5 or 6 members, or by halo-, C₁-C₆alkyl-, C₁-C₆haloalkyl-, C₁-C₆cycloalkyl-, hydroxy-, C₁-C₆alkoxy-, C₁-C₆haloalkoxy- or nitro-substituted CH₂-heteroaryl, wherein the aryl moiety has 5 or 6 members; or which may be substituted by heteroaryl, wherein the aryl moiety has 5 or 6 members, or by halo-, C₁-C₆alkyl-, C₁-C₆haloalkyl-, hydroxy-, C₁-C₆alkoxy-, C₁-C₆haloalkoxy-, cycloalkyl- or nitro-substituted heteroaryl, wherein the aryl moiety has 5 or 6 members; and

which may contain a fused or spiro-bound alkylene or alkenylene chain containing from 2 to 6 carbon atoms, which chain may be interrupted by oxygen or sulfur atoms,

G is hydrogen, -C(X₁)-R₃₀, -C(X₂)-X₃-R₃₁, -C(X₄)-N(R₃₂)-R₃₃, -SO₂-R₃₄, an alkali metal cation, alkaline earth metal cation, sulfonium cation or ammonium cation, -P(X₅)(R₃₅)-R₃₆ or -CH₂X₆C(X₇)-R₃₇, -CH₂X₈C(X₉)-X₁₀-R₃₈, -CH₂X₁₁C(X₁₂)-N(R₃₉)-R₄₀ or -CH₂X₁₃SO₂-R₄₁, wherein X₁, X₂, X₃, X₄, X₅, X₆, X₇, X₈, X₉, X₁₀, X₁₁, X₁₂ and X₁₃ are each independently of the others oxygen or sulfur, and R₃₀, R₃₁, R₃₂, R₃₃, R₃₄, R₃₅, R₃₆, R₃₇, R₃₈, R₃₉, R₄₀ and R₄₁ are each independently of the others hydrogen, C₁-C₁₂alkyl or C₁-C₁₂alkyl substituted by halogen, formyl, cyano, nitro, tri-C₁-C₆alkylsilyl, hydroxy, C₁-C₆alkoxy, C₁-C₆alkoxycarbonyl, amino, C₁-C₆alkylamino, di-C₁-C₆alkylamino, mercapto, C₁-C₆alkylthio, C₁-C₆alkylcarbonyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylcarbonylamino, C₁-C₆alkoxycarbonylamino, C₁-C₆alkyl-

aminocarbonylamino, C₁-C₆alkylthiocarbonyloxy, C₁-C₆alkylthiocarbonylamino, C₁-C₆-alkoxythiocarbonyl, aminothiocarbonyl, C₁-C₆alkylthiocarbonyloxy, C₁-C₆alkylthiocarbonylamino, C₁-C₆alkoxythiocarbonylamino, C₁-C₆alkylsulfinyl, C₁-C₆alkylsulfonyl, C₁-C₆alkylsulfonyloxy, C₁-C₆alkylsulfonylamino, C₁-C₆alkoxyimino, hydroxyimino, heteroaryl, benzyloxy, phenoxy or by halophenoxy; or

C₂-C₁₂alkenyl, C₂-C₁₂alkenyl, C₃-C₁₂cycloalkyl, C₃-C₆cycloalkyl substituted by halogen, C₁-C₆-haloalkyl, C₁-C₆alkyl, C₁-C₆alkoxy, C₁-C₆alkylcarbonyloxy, C₁-C₆thioalkyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, tri-C₁-C₆alkylsilyl or by tri-C₁-C₆alkylsilyloxy; phenyl or phenyl substituted by alkoxy, halogen, C₁-C₆haloalkyl, nitro, cyano, C₁-C₆alkyl, C₁-C₆alkylcarbonyloxy, C₁-C₆thioalkyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, tri-C₁-C₆alkylsilyl or by tri-C₁-C₆alkylsilyloxy; heteroaryl or heteroaryl substituted by halogen, C₁-C₆haloalkyl, nitro, cyano, C₁-C₆alkyl, C₁-C₆alkoxy, C₁-C₆alkylcarbonyloxy, C₁-C₆thioalkyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, tri-C₁-C₆alkylsilyl or by tri-C₁-C₆alkylsilyloxy; and

R₃₄ is additionally C₂-C₂₀alkenyl or C₂-C₂₀alkenyl substituted by halogen, C₁-C₆alkylcarbonyl, C₁-C₆alkoxycarbonyl, C₁-C₆alkylcarbonyloxy, C₁-C₆alkoxy, C₁-C₆thioalkyl, C₁-C₆alkylthiocarbonyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylsulfonyl, C₁-C₆alkylsulfinyl, C₁-C₆alkylaminosulfonyl, di-C₁-C₆alkylaminosulfonyl, C₁-C₆alkylsulfonyloxy, C₁-C₆alkylsulfonylamino, C₁-C₆alkylamino, di-C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, di-C₁-C₆alkylcarbonylamino, cyano, C₃-C₆cycloalkyl, C₃-C₆heterocyclyl, tri-C₁-C₆alkylsilyl, tri-C₁-C₆alkylsilyloxy, phenyl, substituted phenyl, heteroaryl or by substituted heteroaryl; or

C₂-C₂₀alkynyl or C₂-C₂₀alkynyl substituted by halogen, C₁-C₆alkylcarbonyl, C₁-C₆alkoxycarbonyl, C₁-C₆alkylcarbonyloxy, C₁-C₆alkoxy, C₁-C₆thioalkyl, C₁-C₆alkylthiocarbonyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylsulfonyl, C₁-C₆alkylsulfinyl, C₁-C₆alkylaminosulfonyl, di-C₁-C₆alkylaminosulfonyl, C₁-C₆alkylsulfonyloxy, C₁-C₆alkylsulfonylamino, C₁-C₆alkylamino, di-C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, di-C₁-C₆alkylcarbonylamino, cyano, C₃-C₇cycloalkyl, C₃-C₇heterocyclyl, tri-C₁-C₆alkylsilyl, tri-C₁-C₆alkylsilyloxy, phenyl, substituted phenyl, heteroaryl or by substituted heteroaryl; or

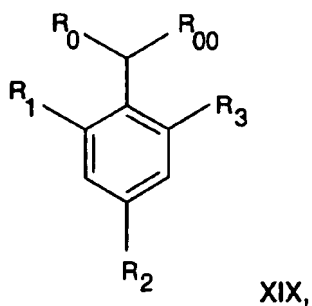
C₃-C₈cycloalkyl or C₃-C₈cycloalkyl substituted by halogen, C₁-C₆haloalkyl, C₁-C₆alkyl, C₁-C₆alkoxy, C₁-C₆alkylcarbonyloxy, C₁-C₆thioalkyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, tri-C₁-C₆alkylsilyl or by tri-C₁-C₆alkylsilyloxy; or heteroaryl or heteroaryl substituted by halogen, C₁-C₆haloalkyl, nitro, cyano, C₁-C₆alkyl,

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C₁-C₆alkoxy, C₁-C₆alkylcarbonyloxy, C₁-C₆thioalkyl, C₁-C₆alkylcarbonylthio, C₁-C₆alkylamino, C₁-C₆alkylcarbonylamino, tri-C₁-C₆alkylsilyl or by tri-C₁-C₆alkylsilyloxy; or heteroaryloxy, substituted heteroaryloxy, heteroarylthio, substituted heteroarylthio, heteroarylamino, substituted heteroarylamino, diheteroarylamino, substituted diheteroarylamino, phenylamino, substituted phenylamino, diphenylamino, substituted diphenylamino, cycloalkylamino, substituted cycloalkylamino, dicycloalkylamino, substituted dicycloalkylamino, cycloalkoxy or substituted cycloalkoxy, or a salt or diastereoisomer of a compound of formula I.

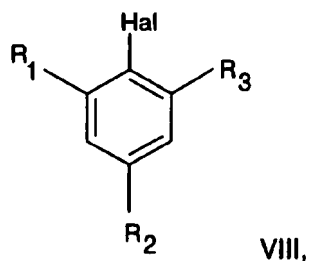
2. A process for the preparation of a compound of formula I according to claim 1, wherein
a) a compound of formula II is reacted with an aromatic zinc or tin compound, an aromatic boric acid, an aromatic boric acid ester or an aromatic Grignard compound, or b) a compound of formula IV or V is reacted with a hydrazine of formula III.
3. A herbicidal and plant-growth-inhibiting composition that comprises a herbicidally effective content of a compound of formula I on an inert carrier.
4. A method of controlling undesired plant growth, wherein a herbicidally effective amount of an active ingredient of formula I or of a composition comprising such an active ingredient is applied to the plants or to the locus thereof.
5. A method of inhibiting plant growth, wherein a herbicidally effective amount of an active ingredient of formula I or of a composition comprising such an active ingredient is applied to the plants or to the locus thereof.
6. A composition according to claim 3 that comprises a further herbicide.
7. A composition according to claim 3 that comprises a safener.
8. A compound of formula XIX

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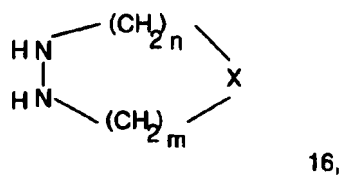
wherein R_0 is OR_7 , OR_8 , NR_7R_9 , NR_8R_{10} or cyano and R_{00} is hydrogen, OR_7 , OR_8 , NR_7R_9 , NR_8R_{10} or cyano, and R_1 , R_2 and R_3 and R_7 to R_{10} are as defined in claim 1, but R_1 and R_3 are not simultaneously hydrogen.

9. A compound of formula VIII



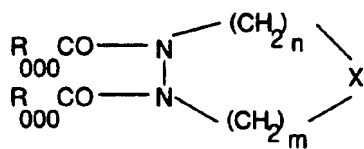
wherein R_1 , R_2 and R_3 are as defined in claim 1, and Hal is chlorine, bromine or iodine, Hal being other than iodine when R_1 and R_3 are methyl and R_2 is phenyl.

10. A process for the preparation of a hydrazine of formula 16



wherein n is 2 or 3, m is 2 or 3 and X is a chemical bond, oxygen or sulfur, in which process a compound of formula 15

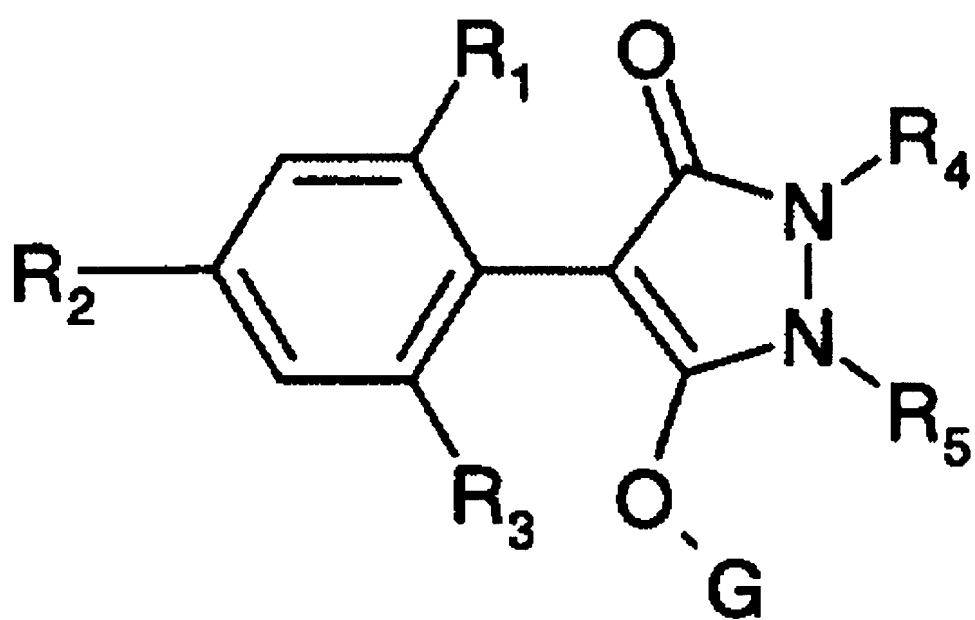
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15,

wherein R_{000} is C_1 - C_4 alkyl and n , m and X are as defined, is reacted in an anhydrous alcohol with an acyl halide or with a hydrohalic acid.

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(I)